#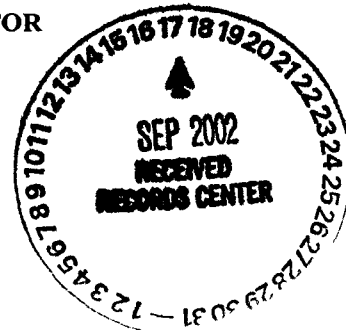 DRAFT FINAL REPORT ON PHASE SPECIATION OF PU AND AM FOR 'ACTINIDE MIGRATION STUDIES AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE'

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August 24, 2000

Significant Findings in FY00

1) **Actinide Phase Speciation** Total ^{239,240}Pu and ^{241}Am concentrations in the storm runoff and pond discharge samples were below the discharge limit of 0 15 pCi/L Both Pu and Am activity concentrations in storm runoff (GS10 on 5/8/00) and pond discharge (GS03 on 4/27/00) samples were also comparable to those determined in 1999 samples from Walnut Creek at GS03 Both storm runoff and pond discharge sampled this year showed again a substantial fraction of Pu and Am in the 0 5 μm filter-passing fraction The majority of Pu and Am in the 0 5μm filter-passing fraction was colloidal, i e , it was filtered out by 100kDa or 3kDa ultrafilters using cross-flow ultrafiltration (CFUF), with only a small fraction of the 0 5μm filter-passing Pu and Am passing a 3 kDa ultrafilter Pu activity concentrations in the particulate fractions (>20 μm & 0 5-20 μm) amounted to about 60-80% of the total

2) **Colloidal Pu remobilization during soil erosion** Pu exceedances ($\geq$ 0 15 pCi/L) occur predominantly during storm runoffs in early spring and summer During these events, elevated concentrations of colloidal Pu, accompanying those in the particle phase, are being observed (Santschi et al , 1999) We hypothesized that colloidal Pu is generated by soil erosion and transport, and that remobilization of colloid-bound Pu during soil erosion events is likely aided by elevated concentrations of humic acids in soil waters During this funding year, we tested this hypothesis in soil erosion and resuspension experiments with RFETS soils, and demonstrated that there is a close relationship between the remobilization of colloidal Pu ($\leq$0 4 μm) and the dissolved organic matter (DOM) concentration (i e , humic acids) in the water at environmentally relevant concentrations We furthermore observed a kinetic effect, i e , an increasing colloidal Pu concentration at increasing resuspension time, and a particle-concentration effect, i e , a decreasing phase partition coefficient of Pu (R_p {in L/kg} = ratio of particulate concentration {in pCi/kg} to the filter-passing concentration {in pCi/L}) with increasing particle concentration (C_p, in kg/L), as $\log(R_p) = 3\,2 - 0\,74 \log(C_p)$ Which of these factors is dominant in the field will require further analysis of field data

3) **Colloid Composition** Isoelectric focusing of ^{14}C radiolabelled colloids showed again that they contain strong acid functional groups which impart the organic fraction a negative charge at natural pH's Bulk colloidal matter contains also strong basic functional groups, as was evident from ^{59}Fe labeling experiments Colloids isolated from the pond discharge and storm runoff samples contained about 3 to 12 % organic carbon, 0 02 to 1 % Fe, and 0 02 to 1 3 % Al, with the higher value for the GS10 sample from 5/8/00 These values agree with their predominantly inorganic nature The clay- and organic carbon - rich nature of the colloids was corroborated by additional TEM and EDAX images, which also suggest that GS10 colloids contain larger amounts of Fe-rich colloids

4) **Particle and Th(IV) residence times in Pond B5** Th(IV)-scavenging and particle residence times in pond B5, derived from measurements of ^{238}U/^{234}Th disequilibria in the water, were of

the order of a few hours to a few days, respectively, supporting the effectiveness of the ponds in reducing particle and Pu loads

1. Overview, Questions and Hypothesis

The goal of the Actinide Migration Evaluation (AME) is to provide data in support of Site closure, including 1) soil action levels vis-à-vis surface water quality, 2) long-term disposition of the ponds, 3) 'far-field' actinide behavior during long-term closure

Four broad areas of investigation emerged from group discussions with AME members and site personnel for consideration for FY 2000 work

1) Phase speciation studies of actinides in surface waters,
2) Colloid characterization studies,
3) Studies of colloid generation during simulated soil erosion (i e , soil resuspension) events,
4) Determination of in-situ removal time studies for particles and colloids in ponds

The following <u>question</u> was addressed in the FY2000 research What causes exceedances of Pu and Am concentrations in RFETS surface waters, which often are associated with storm runoff?

The following <u>Hypothesis</u> were tested in the FY2000 research The exceedances are related, besides hydrodynamic conditions (i e , storm surface runoff vs base flow), to the nature and types of organic matter present in soils, surface and pond water at the time of sampling Organic matter can modify the surface charge and characteristics of particle and colloid aggregates On the one hand, humic and fulvic acids can impart a negative surface charge to particles and colloids, which can promote disaggregation and dispersion of aggregates, and thus, increased mobility and concentrations of colloidal species in surface waters On the other hand, large, surface-active, organic molecules such as exopolymeric acid polysaccharides from bacteria and algae act to bind colloidal and particulate species together, and thus, cause their removal and lower their concentrations in surface waters

2. Justification

A wide range of work at Rocky Flats has demonstrated that 'particulate' and 'colloidal' forms of Pu and Am make up a significant fraction of actinides in surface waters and suggests that surface water transport of Pu and Am is dominated by aggregation and disaggregation processes of particles and colloids during soil erosion events A clear understanding of the phase speciation of Pu is a prerequisite for the development of defensible closure strategies Questions concerning the environmental form of Pu and Am have direct bearing on evaluating the importance of various migration pathways and in the development and refinement of Data Quality Objectives (DQO i e , model parameters and uncertainty levels)

3. Tasks

3.1 Task 1. Colloid and particulate associations of Pu and Am

In FY2000 we continued the phase speciation work of Pu and Am in stream water, using the sites already investigated in 1998 and 1999

3 1 1 Objectives

1   To determine the association of Pu and Am with 1) particulate, 2) colloidal and 3) dissolved phases for field samples from a selected pond, pond release waters and a future compliance point
2   To determine if natural organic matter plays a role in the formation of colloids that facilitate transport of actinides from contaminated soil source terms
3   To determine the chemical nature of the colloidal carrier phase (e g , Fe, Mn, C, Al, etc)

3 1 2 Justification

FY 98&99 work has demonstrated that 0 5 μm filter-passing Pu is predominantly in a colloidal form   The issue of the speciation of filter-passing forms of Pu is crucial for understanding of transport and bioavailability, i e , the relationship between surface water quality and soil action levels, as well as to support the scientific defensibility of RESRAD model simulations   The distribution of Pu and Am among different particles sizes and colloid molecular weights is important for developing management controls on surface water quality

3 1 3 Analytical Plan

Phase association studies of Pu and Am investigations involved the following subtasks

a)   Surface water sampling by CFUF,
b)   Colloid isolation and characterization,

The selected sampling sites were pond B5, pond release waters at a compliance point (GS03), and G10 storm runoff (Figure 1)   Grab samples were collected by bailing water from the stream using a small container followed by compositing the water into clean 15 to 20 Liter Nalgene carboys for processing and analysis in the laboratory (Figure 2)   In addition, soil samples were collected for extraction of natural organic matter fractions and used in the soil erosion/colloid stability experiments at TAMUG

The methods for isotope separation were adapted from EPA Method 908 0 (1980), USDOE (1979), USEPA (1979), and Yamato (1982) as described also in (Appendix 2). Each sample was acidified with concentrated nitric acid to pH <2 and allowed to sit for at least 16 hours   For each sample concentrated hydrochloric acid was added at 5ml/L and ^{243}Am and ^{242}Pu yield tracers were added   The samples were placed on a stir plate and 5 ml of 40 mg/ml Fe(III) carrier was added   The pH was measured and concentrated hydrochloric acid added until pH is <1   The sample was covered and stirred for 30 minutes and the pH measured again

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Once the pH was <1, concentrated ammonium hydroxide was added until turbidity remained constant then an additional 50 mls was added. The sample was again covered and stirred. After 30 minutes, the sample was removed from the stir plate, the stir bar removed and the precipitate was allowed to settle. The supernate was decanted until the precipitate slurry could be transferred to 250 ml centrifuge tubes. The samples were centrifuged for 30 minutes at 3000 rpm. The supernate was decanted and the precipitate was shipped to Texas A&M University. Once at Texas A&M University, the samples were dissolved in concentrated HCl to which 75 mls of 9 N HCl and 2 ml saturated sodium nitrite were added. The samples were then run through a series of three anion exchange columns (Appendix 2). The first column separated the Am from the Pu fractions. The Pu was then microprecipitated on a filter, mounted on a stainless steel planchet and alpha counted. The Am fraction was carried through a methanolic anion exchange column followed by a TEVA resin column. The Am fraction was microprecipitated, mounted on a stainless steel planchette and alpha counted.

The protocols of Guo and Santschi (1996,1997) and Wen *et al* (1996, 1999) were followed for isolating colloidal and particulate phases of metals such as Pu, Am from surface waters by CFUF. Chemical parameters measured in the aqueous sample fractions were
- total organic carbon (TOC),
- dissolved organic carbon (DOC),
- colloidal organic carbon (COC),
- particulate organic carbon (POC),
- pH,
- alkalinity, and
- Al, Fe and Mn

Chemical parameters to be measured in the filtered solids (colloid and particulate phases) are
- % organic carbon,
- Al, Fe and Mn

Analytical methods followed the protocols of Guo and Santschi (1997) and Wen *et al* (1999). These measurements were conducted on the isolated aqueous solution phases or on resolubilized freeze dried material.

Aliquots of colloid and particle samples were kept for TEM imaging analyses. These analyses were used to visually identify the gross composition of the particulate and colloidal phases.

In selected cases, electrophoretic focusing experiments (Quigley, 2000) determined isoelectric points of isolated colloids.

A note on terminology We denote all fractions with either the upper or lower size or nominal molecular weight cutoff limit, or both. The terms "dissolved", "filtrate" are ambiguous, and the terms "retentate" and "permeate" are reserved for fractions which were retained by or had permeated an ultrafilter.

3 1 4   Strategy

Surface water samples from the field were taken in an extended sampling expedition in April/May 2000. Laboratory processing of samples was carried out in the following months

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3.2 Task 2: Soil erosion/Colloidal Pu release experiments

The laboratory experiments involved treatment of actinide-contaminated soil with extracted, natural organic matter to investigate natural mechanisms of colloid formation during storm water runoff

3 2 1 Objectives

1 To determine if natural organic matter plays a role in the formation of colloids that facilitate transport of actinides from contaminated soil source terms
2 To experimentally determine what natural processes favor particle and colloid stabilization or destabilization using experimental approaches given in Wilkinson et al (1997)

3 2 2 Justification

A better understanding of the process(es) which control the speciation of filter-passing forms of Pu is crucial for evaluating their transport and bioavailability A major focus of this task is to determine the dominant Pu species generated during soil resuspension during storm runoff Some of the major processes which control colloid formation will be studied in controlled laboratory experiments, with the purpose of relating colloid stability to organic matter composition The type of natural organic matter can control the tendency of particles and colloids to aggregate For example, small organic molecules such as fulvic acid and other organic acids can increase dispersion of soil colloids through their effect on particle charge, while large, surface-active, organic molecules such as polysaccharides act to bind colloid particles together (e g , Gu and Doner, 1993, Kretzschmar et al , 1997, Wilkinson et al , 1997) Colloid stability is an important colloid characteristic because it reflects the extent to which colloidal material interacts with particles and immobile soil media. Therefore, colloid stability is a primary parameter for 1) estimating colloid mobility and for 2) development of engineering and management strategies for removing colloidal-associated actinides from surface water

3 2 3 Analytical Plan

Colloid formation and associated actinide partitioning was investigated in controlled soil erosion/resuspension experiments, carried out with variable amounts of commercial organic matter compounds (i e , Aldrich humic acids, exopolymers of microorganisms, such as Alginic Acids, Xanthen) and soil extracts (e g , humic acids) Pu -contaminated soils from the Site were resuspended in the presence or absence of these natural organic substances, and separated solutions were processed as described above to determine if Pu colloid formation occurs The colloids in solution was analyzed for DOC, Pu and Am to determine whether colloid formation also liberated Pu and Am from the soils A schematic of these experiments is displayed in Figure 3 Pu-contaminated soil in 0 5µm filtered tap water served as a control

3 2 4 Strategy

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The strategy was to carry out these soil erosion/resuspension and colloid remobilization experiments using existing samples Laboratory processing of these samples was carried out throughout the year

3.3 Task 3: In-situ particle and colloid residence time determination

We determined in-situ particle and colloid residence times in pond waters using the $^{234}Th/^{238}U$ disequilibrium technique (e g , Baskaran et al , 1996, Guo et al , 1997, Santschi et al , 1999a)

3 3 1 Objective

To investigate if in-situ particle and Th(IV) residence time determination in pond waters using the $^{234}Th/^{238}U$ disequilibrium technique prove to be feasible

3.3 2 Justification

FY 98&99 work has demonstrated that 0 5 μm filter-passing Pu is predominantly in a colloidal form. Colloid stability is a primary parameter for 1) estimating colloid mobility and for 2) development of engineering and management strategies for removing colloidal-associated actinides from surface water Particle and colloids residence times in ponds are likely short, as actinides are removed to better than 90% in the pond water in about 45 days, according to Site sources Better knowledge of residence times of particles and colloids would facilitate a future decision about pond operations

3 3.3 Analytical Plan

Because ^{238}U concentrations was high enough (i e , 1 dpm/l, or higher), and relatively constant over time, the $^{234}Th/^{238}U$ disequilibrium technique (e g , Baskaran et al , 1996, Guo et al , 1997, Santschi et al , 1999a), it can provides estimates of particle and colloid residence times at steady state from the deficiency of ^{234}Th with respect to its mother nuclide, ^{238}U, in particle, colloid and solution phases Large volumes of water were pumped from the ponds through one prefilter and two MnO_2 impregnated fiber filters, which efficiently extract ^{234}Th The efficiency of extraction is determined from the activity ratio in the two cartridges, and is usually over 90% (Baskaran et al., 1993)

3 3 4 Strategy

Surface water samples from pond B5 were taken in a sampling expedition in April 26-27 by pumping water through extractor cartridges in the field Laboratory processing of samples was carried out in the month following sampling

4. RESULTS AND DISCUSSION

During this quarter we sampled, filtered, ultra-filtered and processed a discharge sample on 4/27/00 and a storm runoff sample on 5/8/00, in duplicate For the discharge sample,

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approximately 200 liters of water was taken from GS03 and brought in various containers to CSM, where it was combined in a 55 gallon drum The 200 liters of storm runoff were collected from GS10 and brought to CSM in two 55 gallon drums Both samples were size fractionated in duplicate for a total of 4 samples of approximately 90 liters each 12 - 18 liters of total water (unfiltered) was set aside for each sample The remaining water was passed through 20 micron and 0 5 micron filters and collected in 5 gallon carboys One carboy of 0 5 micron filtered water was set aside for later analysis of the 0 5 micron filter passing fraction. The remaining 0 5 micron filtered water was used for the cross flow ultrafiltration (CFUF) Thirty to forty liters were run through a 3 kDa filter membrane and another thirty to forty liters were run through a 100 kDa filter membrane All size fractions were processed by $Fe(OH)_3$ precipitation at CSM The resultant precipitate was sent to TAMUG for radiochemical analysis of ^{239,240}Pu and ^{243}Am

In a another experiment, Pond B5 was sampled 4/26/00 and 4/27/00 for ^{234}Th and ^{238}U to calculate residence times of the particulate and filter-passing phases Th-234 was extracted by pumping hundreds of liters of water through a 0 5 μm prefilter and two 0 5 μm MnO_2 impregnated cartridge filters Filters were then combusted in a muffle furnace and the ash was packed into counting vials and counted on a well Ge gamma detector ^{238}U was determined by ICP-MS

At TAMUG, we also conducted soil erosion/resuspension experiments using Rocky Flats soil (99A3372-002 006) as a source of Pu Soil concentrations, resuspension times, as well as type and quantity of organic substances were varied to assess their effects on the <0 45 μm filter-passing fraction ^{239,240}Pu activities

4.1. Pu and Am concentration and phase speciation data from pond and storm discharge samples:

^{239,240}Pu and ^{241}Am phase speciation results are shown in Tables 1 and 2, and ancillary results in Tables 3-11 Unfortunately, in the first pond discharge sample A, activities Pu and Am activities in the 3kDa ultrafiltration samples were anomalous The problem was attributed to the tracer not having equilibrated with the sample, likely due to interference by organics To alleviate the organic matter problem, all subsequent samples (pond discharge B and storm runoff A and B) were treated with strong acids (HCl and HNO_3) first, and evaporated after tracer addition prior to the column chemistry The second pond discharge sample B, then gave more reliable results, as judged from the activity balance

As had been observed in previous years, most of the Pu and Am in the water (Table 1, Figures 4-5) was found in the particulate phase, with most of the remainder in the colloidal phase Pu/Am ratios (Table 12) of the different phases and samples indicate that the storm runoff samples generally had lower ratios than the pond discharge samples Values of phase partition coefficients, Rp, range from 4 8 to 8 0 10^4 L/kg (Table 13)

4.2. Colloid characterization:

ICP-MS, CHN, isoelectric focusing and SEM were used for characterizing freeze-dried colloids from the April and May sampling expeditions Results are given in Table 13 As in previous years, colloids were primarily composed of clay and organic matter colloids (0 02-1 % of Fe, 0 02-1 3 % of Al, 3 5-11 3 % of OC, see Tables 8, 9) This clay - rich colloid matrix was further confirmed using electron microscopy techniques

Colloids were radioactively tagged using ^{14}C and ^{59}Fe, using protocols given in Santschi et al (1999), and their isoelectric points determined in a gel electrophoresis (Figure 6, Table 14)

revealed components with strong acid functional groups (labeled by ^{14}C on the C-OH sites) and basic functional groups (labeled by ^{59}Fe)

Transmission Electron Microscopy (TEM) combined with Enery Dispersive X-Ray Microprobe Analysis (EDAX) Aliquots of freeze-dried colloids (RF137 and RF157) were analyzed by Marcia West and Gary Leppard at McMaster University Major findings The main colloids in both samples were clays (some of which contained Fe), microbes (mainly bacteria), microbe parts, debris, biogenic silicates, membranous structures (probably biological), titanium microcrystals and extremely small colloids aggregated into complex structures These latter aggregates of extremely small colloids could not be properly analyzed morphologically because the samples had been processed as dehydrated powders Thus the fractal arrangements and long range associations would almost certainly have been artifactually altered (see earlier work of Buffle and Leppard) The samples were infiltrated and embedded in Nanoplast FB101 melamine resin, then sectioned. The ultra-thin sections (60-80 nm) were made on a Reichert Ultracut E microtome, then collected on uncoated 200-mesh Cu grids for morphological characterization and on Formvar, Carbon-coated 100-mesh Ni grids for EDS analysis The sections were viewed in a JEOL JEM 1200 EX TEMSCAN microscope operating at 80kV The range of primary magnifications used for morphological analyses was 10,000-75,000 X The microscope is equipped with a Tracor Northern EDS detector (laboratory resolution of 150 7 eV), connected to a computer that runs Iridium System (Version 2 2) software (IXRF Systems, Inc) The duration for each point of analysis was 100s with parameters such as kV, tilt, spot size and working distance kept the same for all analyses The spectra obtained are keyed to individual particles on micrographs some of which are shown in Figures 7 and 8

The freeze-drying method that was employed clumps the colloids severely, making fine distinctions between the two samples difficult to ascertain Perhaps sample RF157 was more mineralized than sample RF137, which is corroborated by the metal analysis of the colloid species (Table 12) Sample RF157 was richer in Fe-containing clays than was RF137 (Figures 7 and 8), in agreement with the analytical data (Table 12) For morphological analyses, in the future we consider preparing the samples in Nanoplast on site with no sample storage whatsoever, possibly as a part of a multi-method protocol employing some conventional methods for TEM sample preparation While Nanoplast preserves the three-dimensional aspect of colloid aggregates, it does not provide good contrast for cells or organic materials in general A second protocol used correlatively, could correct for this problem, such a protocol will be applied at the time of FY2001 sampling

4.2. $^{238}U/^{234}Th$ disequilibrium to derive thorium scavenging and particle residence times in pond B5:

Preliminary results indicate that the ^{238}U concentrations in pond B5 were 2 28±0 04 µg/L (0 76±0 01 pCi/L) in unfiltered water, 2 11±0 11 µg/L (0 71±0 04 pCi/L in 0 5 µm filtered water, the ^{234}Th concentration in the 0 5 µm filter-passing fraction was equal to or less than 0 005 pCi/L, three times our detection limit, while the particulate (≥0 5 µm) $^{234}Th_p$ was about 0 04 pCi/L or less

From these activity determinations, the scavenging and particle residence times (τ_i) can be determined, as follows

$\tau_i = \tau_{234} \times ([^{234}Th_i]/[^{238}U])$,

where i = total, dissolved, or particulate (Baskaran et al , 1996, Santschi et al , 1999)

With $\tau_{234} = \lambda_{234}^{-1}$, λ_{234} = decay constant of ^{234}Th = 0 03 d^{-1}, $[^{234}Th_i]/[^{238}U_i]$ = activity ratio

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Removal residence times of ^{234}Th from the water and particle residence times of ≤ 6 hours and ≤2 days, respectively, could thus be calculated from our measured ^{238}U and ^{234}Th activities in the water These residence times confirm the high efficiency of the holding ponds for particles, and particle-reactive elements such as Pu

4.3. Colloidal Pu release potential from soil erosion/resuspension experiments:

Soil erosion/resuspension experiments with the fine fraction of RFETS soil (719 pCi/g, 4 9% of OC), with 0 5 g of this soil in 190 ml of filtered tap water demonstrate that Pu that is released during these experiments is mostly of colloidal nature (Table 15) As shown in Figures 9 and 10, colloidal Pu concentrations increase over a resuspension time of 1-7 days, in the presence and absence of additional humic acid (at 30 mg DOM/L = 12 mg-C/L of DOC) Thus, there is a significant kinetic effect during the release of colloidal Pu When the results from the control experiment are compared with those with increasing Aldrich humic acid (41% OC) concentrations, it becomes clear that colloidal Pu concentrations significantly increase with increasing humic acid concentrations When soil humic acids from uncontaminated Rocky Flats soils were used (using the procedure given in appendix 3, resulting in16% of OC), they turned out to be considerably more effective than the commercial Aldrich humic acids in remobilizing Pu from contaminated soils into the water, when concentrations are expressed in dissolved organic carbon, DOC, rather than dissolved organic matter (Figures 11 and 12) Please note that DOC concentrations in the water, and organic carbon of suspended matter in the field can be as high or higher than was used in these lab experiments with RFETS soil containing a more refractory form of Pu

There was only a small difference between the control and alginic acid (Figure 13), regardless of the concentration range, when similar resuspension times were compared Soil resuspension experiments with the bacteria-derived acid polysaccharide Xanthen, revealed that it acts as a particle-glue, i e , it exhibited strong particle aggregation effects (e g , strong filter clogging), but showed only insignificant effects on the colloid production potential of resuspended soils, consistent with its particle aggregating effects Its strongest effects is thus on increasing the cohesiveness of soil particles When results at different particle concentrations are compared, it is apparent that Pu remobilization is inversely proportional to the soil particle-water ratio (Figure 14) The ratio of resuspended soil to water is thus an important parameter in predicting Pu colloid formation The phase partitioning coefficient (Rp {in L/kg} = ratio of particulate concentration {in pCi/kg} to the filter-passing concentration {in pCi/L}) for Pu was inversely proportional to particle concentration (Cp, in kg/L), with $Rp = 10^{3\ 2}\ Cp^{-0\ 74}$, or $log(Rp) = 3\ 2 - 0\ 74\ log(Cp)$ (Figure 14) The observation of Rp being a function of particle concentration is consistent with the presence of colloidal Pu in the filter-passing fraction, and is called "particle concentration effect" This phenomenon was previously ascribed to the presence of colloidal species of the analyte, e g , for Th(IV) and other particle-reactive metals (e g , Honeyman and Santschi, 1988, 1989) It suggests that higher colloidal Pu concentrations are also found at times of greater amounts of resuspended soils in the water Even though such an effect might be caused by an organic matter sub-fraction, this particle-concentration effect is much smaller for organic matter itself (Figure 15)

The particle-concentration-effect is, however, also evident from the field samples we collected in1998 to 2000 (including data from Santschi et al , 1998, 1999b, see Figure 14) Field-

derived Rp values for Pu in GS10 and GS03 waters are, however, lower at equivalent suspended matter concentrations, possibly due to differences in organic matter content and/or physical forms of Pu (particle size distribution) It suggests that Pu in soils further away from source areas are in a more "weathered form", i.e., they are associated with finer particles and colloids As a consequence, colloidal fractions of Pu in field samples amount to 30-70 % of the total Pu in the field samples, at suspended matter concentrations (Cp) of 30 - 120 mg/L, with the remainder in the particulate fraction Despite the lowering of the Rp value with increasing Cp concentration, only 0 1 - 1 % of the Pu was remobilized from the soil the lab experiments using RFETS soil taken near the 903 Pad (Figure 16) The same was true for the effects of increasing DOC concentrations

While it is still true that most of the Pu exceedances are produced by high suspended matter concentrations in the water, which contain 0 1-1 pCi/g of Pu, a much greater proportion of Pu in samples from GS03 and GS10 is in a colloidal rather than particulate form As mentioned before, this is likely due to more weathered forms of Pu further away from sources, and more particulate forms of PuO_2 near the 903 Pad, the site where the soil for the laboratory experiments was taken It is also likely that in the field, the quality (i e, composition, e g, humic acids vs aquagenic organic matter such as acid polysaccharides), in addition to the quantity of DOC, will play an important role in remobilizing Pu from soils

Regardless, soil resuspension experiments suggest that colloidal Pu release during soil erosion events increases as a function of

   a) suspended particulate matter concentration,
   b) resuspension time, and
   c) humic acid (DOC) concentrations

Which one of these is the dominant factor in the field will require further analysis of field data.

Pu release from resuspended particles slightly decreases as a function of alginic acid concentrations in the water Alginic acid, an excretion product by both algae and bacteria in soil and water, has an aggregating effect on particles, while humic acids, the main degradation product of plant matter in soils, generally has a dispersing effect

These results thus suggest that the addition of colloidal Pu from sediment resuspension and soil erosion to stream water during pond discharge and storm run-off could increase the total Pu concentration, over that of particulate Pu alone, by a factor of 2-4 The increase depends on the amount of eroded soil particles in suspension, the length of time the soil is resuspended, as well as the humic acid concentration of the soil This conclusion is in agreement with all the field data, which show a good correlation between total Pu concentrations and TSS (total suspended solids) in the water at most measuring stations Scatter around the least squares line is approximately around a factor of 2-4, which can be produced by having colloidal Pu, in addition to particulate Pu, in the water rather than only variable Pu concentrations in suspended matter

4 4. Implications of soil erosion studies Importance of organic matter for Pu remobilization.

One of the conclusions of the soil resuspension experiments is that DOC concentrations have a major effect in remobilizing colloidal Pu into solution DOC concentrations measured in 1998, 1999 and 2000 stream samples varied from 4 to13 mg C/L, with no obvious correlation with Pu concentrations, however This could, however, have been due to compositional differences of DOM between spring and summer, aquagenic vs pedogenic organic matter Furthermore, DOC

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concentrations in soil solutions would still have an indirect effect on the extent of soil aggregation/disaggregation, and thus, on Pu remobilization

The recently described inverse relationship between DOC (i e , humic acid) concentrations and concentrations of major ions (and Si) in streams in a catchment at Sleepers River Research Watershed, Danville, Vermont (Kendall et al , 1999), deserves serious consideration as an explanation of the relationship between humic acid concentrations in streams and major ion composition. The streamwater composition in spring before and after snow melt was explained by two endmembers a) A deep flow path component –geomorphic hollow water or near-stream till water (high in base cations and silica and low in DOC, i e , humic acids), and b) a shallow flow path component – overland flow water or perched soil water (low in base cations and silica and high in DOC, i e , humic acids) Subsurface hillslope waters were not viable end members during snow melt.

They attributed the inverse relationship of DOC (humic acid) with base cation (and Si) concentrations to a mixing of near-stream goundwater and shallow organic horizon flow during peak flow periods The organic horizon flow, however, was transient, since this zone rapidly drained following peak flow events At baseflow, the organic horizon end member did not contribute significantly to streamflow

Kendall et al (1999) furthermore found a relationship between DOC (humic acid), base cations and hydraulic head or water table They called this the "transmissivity feedback hypothesis" Their observations of stream DOC (humic acid) concentrations increasing during high flows, during times when DOC (humic acid) was flushed from shallow soil horizons as groundwater levels rose into those horizons, were thus consistent with their "transmissivity feedback hypothesis"

A similar situation could also occur at the Rocky Flats Environmental Technology Site Thus, we envision the following scenario which would not only explain the Pu exceedances during spring and summer storm events, which are accompanied not only by high particulate, but also high colloidal Pu concentrations

During storm runoff at shallow water tables, DOC (i e , humic acid) and colloidal Pu concentrations would thus be expected to be high (and base cations low), while during runoff events into dry soil with a deep ground water table, DOC and colloidal Pu concentrations would be expected to be lower at that time This aspect might require further study

5. Conclusions

1 Pond and storm discharge samples collected in spring 2000 showed low Pu concentrations, with a large fraction of the Pu (60-80 %) associated with suspended particulate matter, and the 0 5 μm filter-passing fraction of Pu being mostly in colloidal forms Clay- and organic matter - rich colloids are present in different sizes, as was evident from elemental analyses and TEM and EDAX images

2 Scavenging and particle residence times in pond B5, derived from measurements of $^{238}U/^{234}Th$ disequilibria in the water, were of the order of a less than 4 hours to less than 2 days, respectively, supporting the effectiveness of the ponds in reducing particle and Pu loads

3 Soil resuspension experiments thus suggest that Pu release during soil erosion events increases as a function of a) suspended particulate matter concentration, b) resuspension

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time, and c) humic acid (DOC) concentrations Pu release from resuspended particles slightly decreases as a function of alginic acid (an excretion product of algae and bacteria) concentrations in the water Humic acids extracted from Rocky Flats soils were considerably more effective than commercially available Aldrich Humic Acid in remobilizing colloidal Pu from Rocky Flats soils into the water These results suggest that higher colloidal Pu found in stream water during pond discharge and storm run-off could be regulated by the length of time the soil is resuspended, as well as the humic acid concentration of the soil Soil resuspension experiments with the bacteria-derived acid polysaccharide Xanthen may act as a particle-glue, i e , exhibited strong particle aggregation effects (e g , strong filter clogging), but showed only insignificant effects on the colloid production potential of resuspended soils, consistent with its more aggregating nature Its strongest effects is thus in increasing the effective particle size of aggregates, thus reducing soil erosion, rather than in reducing the colloidal Pu production of already resuspended soil

4   The ratio of resuspended soil to water is also important in regulating Pu colloid formation The phase partitioning coefficient, R_p (in L/kg), was inversely proportional to particle concentration (C_p, in kg/L), with $R_p = 10^{3.2} C_p^{-0.74}$, or $\log(R_p) = 3\,2 - 0\,74\,\log(C_p)$ Such a relationship has been described before for Th(IV) (e g , Honeyman and Santschi, 1988, 1989), and is a consequence of the presence of colloidal forms of Pu in the filter-passing fraction

5   A better understanding of the causes for colloidal Pu remobilization during storm runoff will allow better remedial actions to be undertaken after site closure (e g , holding ponds, drainages, etc)
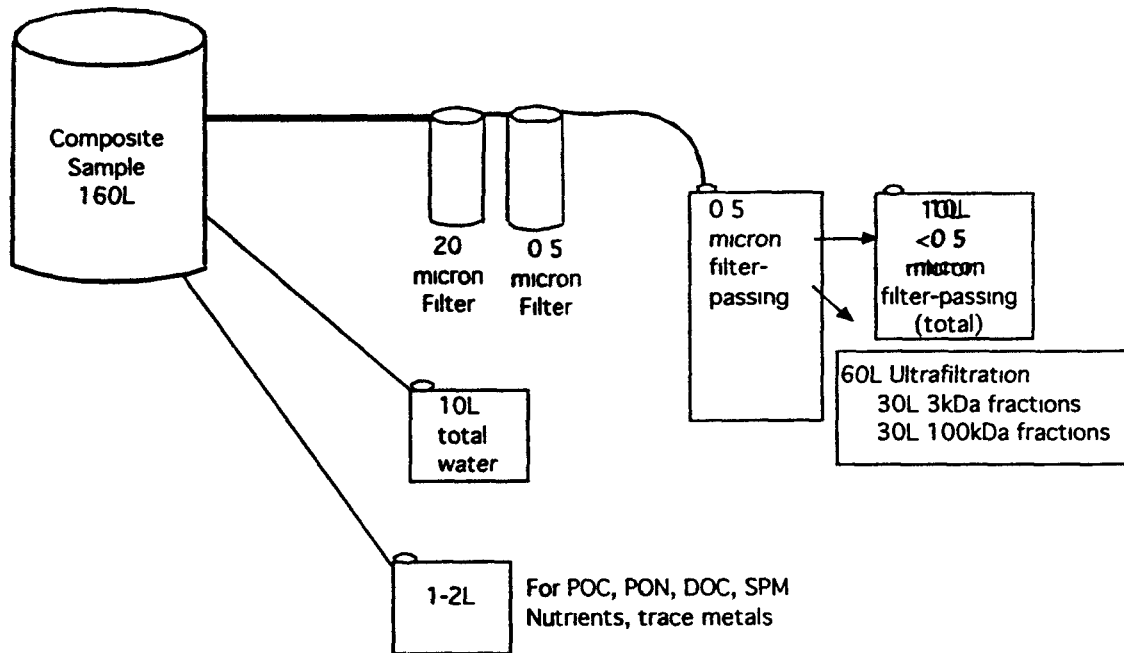

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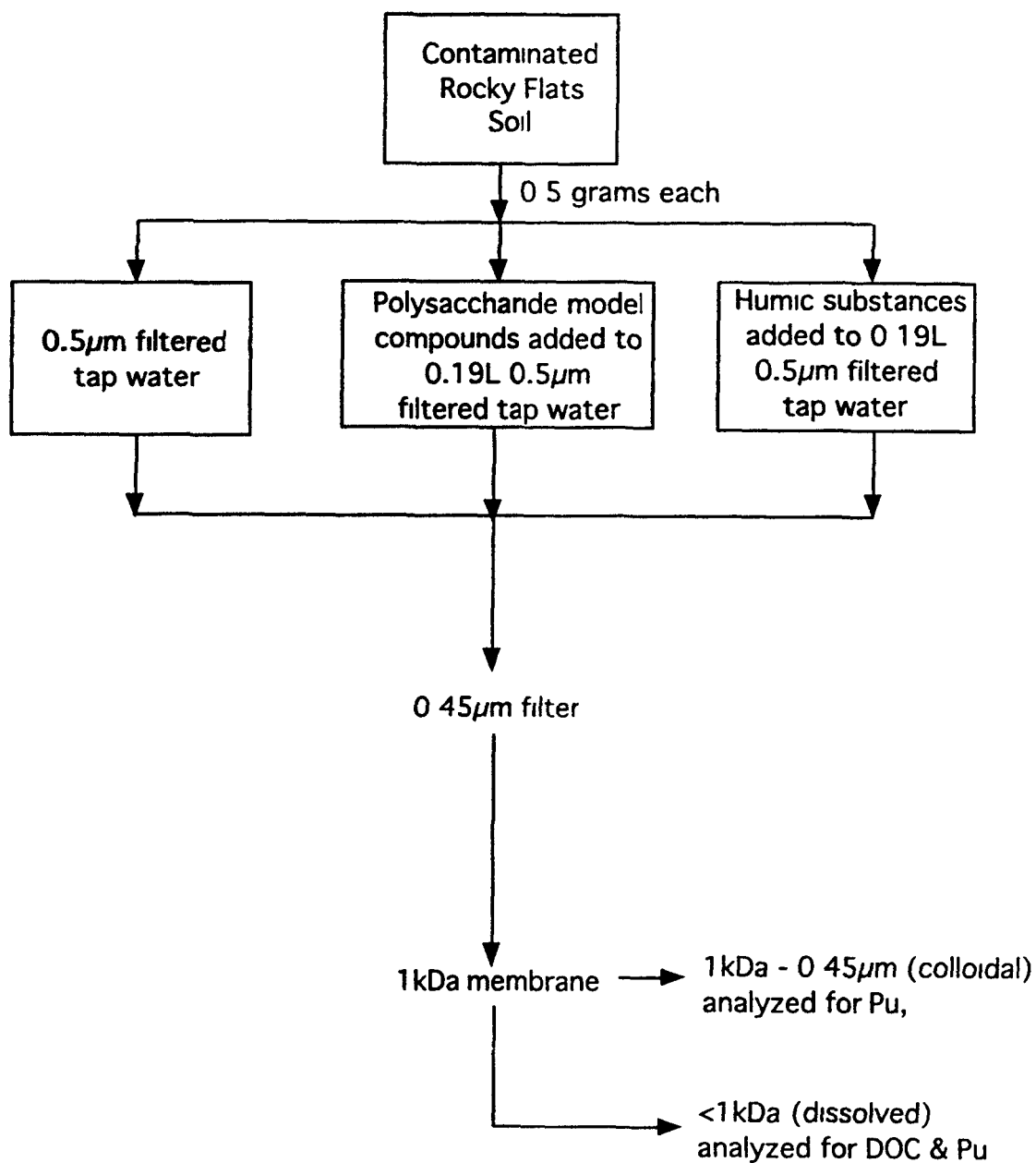
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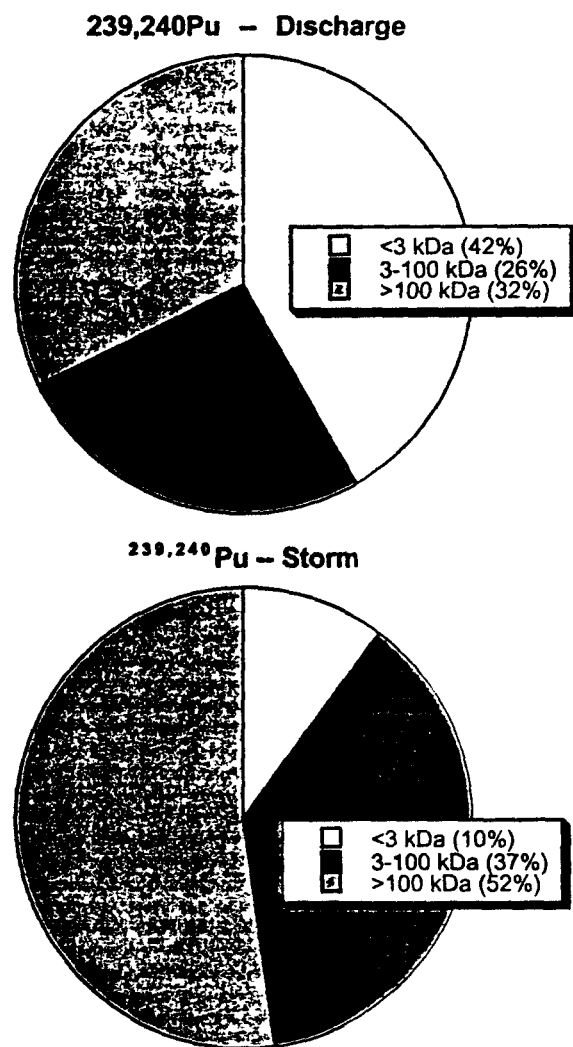
Sampling Setup
(all fractions run in duplicate)

Composite
Sample
160L

20
micron
Filter

0 5
micron
Filter

0 5
micron
filter-
passing

10L
<0 5
micron
filter-passing
(total)

60L Ultrafiltration
    30L 3kDa fractions
    30L 100kDa fractions

10L
total
water

1-2L

For POC, PON, DOC, SPM
Nutrients, trace metals

Figure 2. Schematic of field sampling.

14

Resuspension Experiments

```
                    ┌─────────────────┐
                    │  Contaminated   │
                    │   Rocky Flats   │
                    │      Soil       │
                    └─────────────────┘
                             │ 0 5 grams each
          ┌──────────────────┼──────────────────┐
          ▼                  ▼                   ▼
┌──────────────┐  ┌────────────────────┐  ┌──────────────────┐
│              │  │ Polysacchande model│  │ Humic substances │
│ 0.5μm filtered│ │ compounds added to │  │ added to 0 19L   │
│  tap water   │  │    0.19L 0.5μm     │  │  0.5μm filtered  │
│              │  │  filtered tap water│  │   tap water      │
└──────────────┘  └────────────────────┘  └──────────────────┘
          │                  │                   │
          └──────────────────┼───────────────────┘
                             ▼
                      0 45μm filter
                             │
                             ▼
          1kDa membrane ───────► 1kDa - 0 45μm (colloidal)
                 │                analyzed for Pu,
                 │
                 └───────────► <1kDa (dissolved)
                                analyzed for DOC & Pu
```

Figure 3. Schematic of preliminary soil resuspension experiments Ultrafiltration was only carried out in selected samples.

16

239,240Pu — Discharge

☐ <3 kDa (42%)
■ 3-100 kDa (26%)
▨ >100 kDa (32%)

239,240Pu — Storm

☐ <3 kDa (10%)
■ 3-100 kDa (37%)
▨ >100 kDa (52%)

Figure 4. Partitioning of [239,240]Pu between dissolved and colloidal fractions in the 0 5μm filter-passing fraction.

17

16

²⁴¹ Am – Discharge

<3 kDa (24%)
3-100 kDa (20%)
>100 kDa (56%)

²⁴¹ Am – Storm

<3 kDa (14%)
3-100 kDa (41%)
>100 kDa (45%)
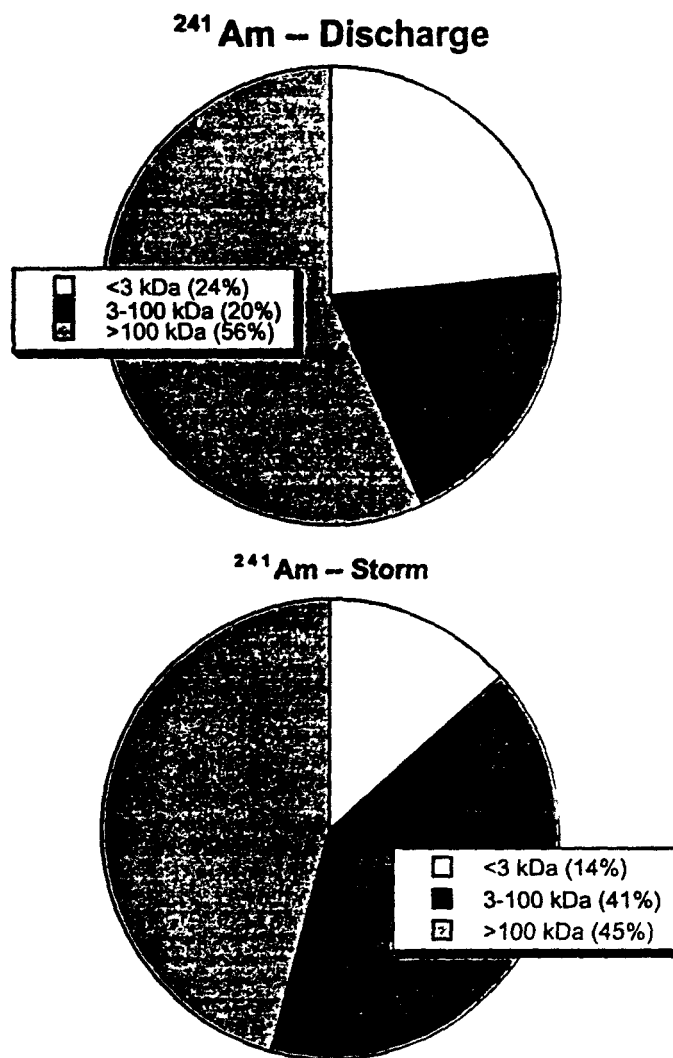
Figure 5. Partitioning of ²⁴¹Am between dissolved and colloidal fractions in the 0.5μm filter-passing fraction.

18

17
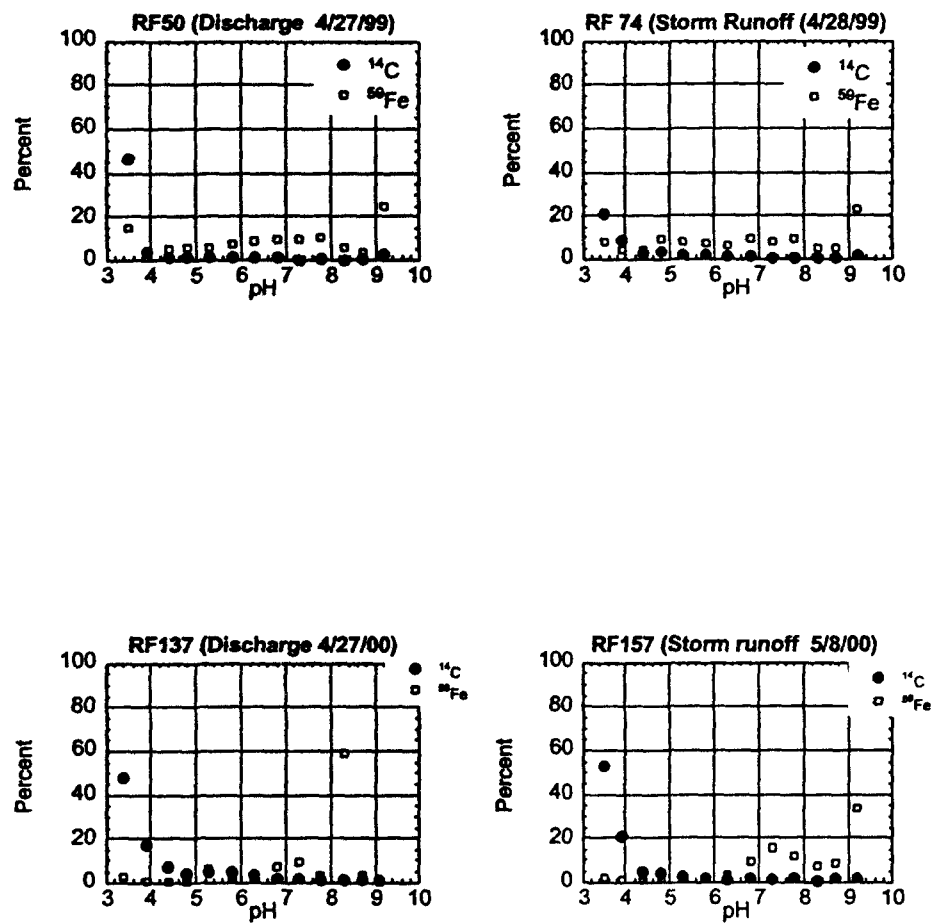
Figure 6. Gel electrophoresis results of colloids taken in 2000 with those taken in 1999.

Figure 7a. TEM pictures of colloid samples. RF137N-1

Figure 7b. TEM pictures of colloid samples. RF137N-2

19
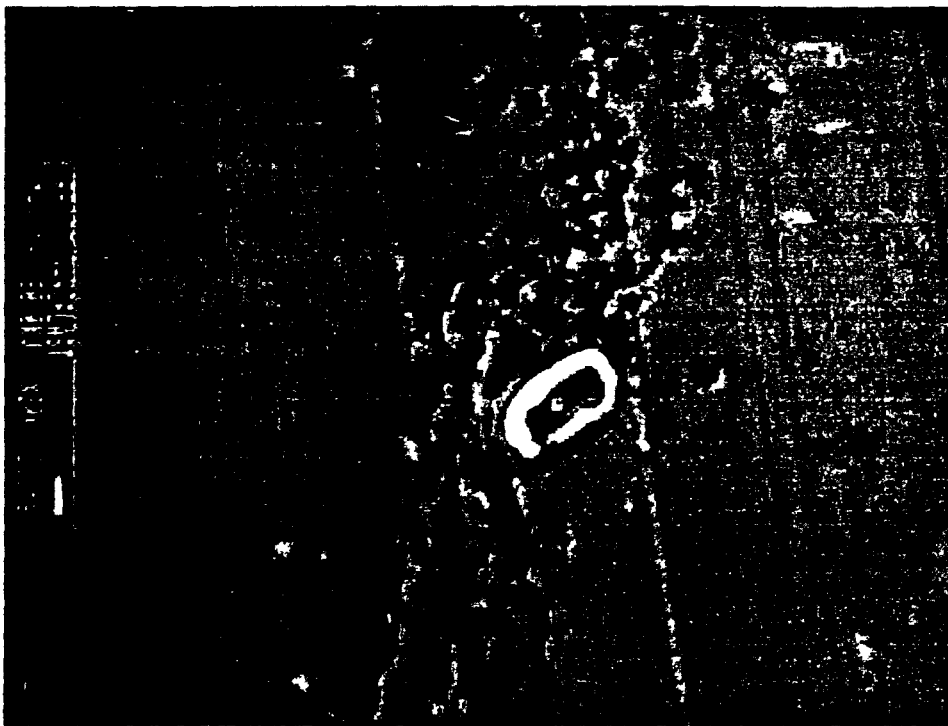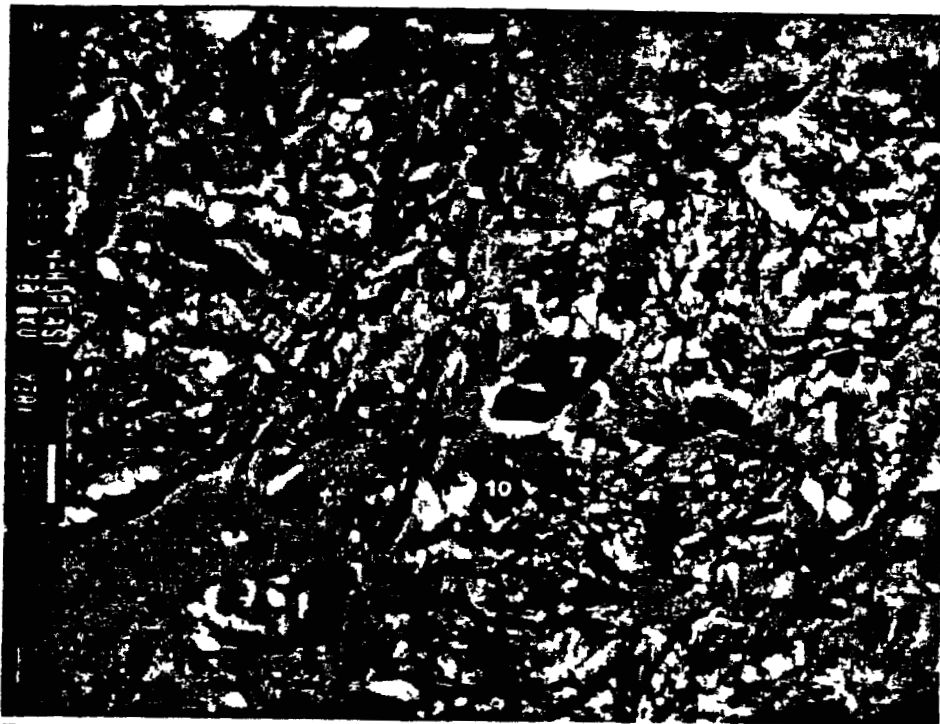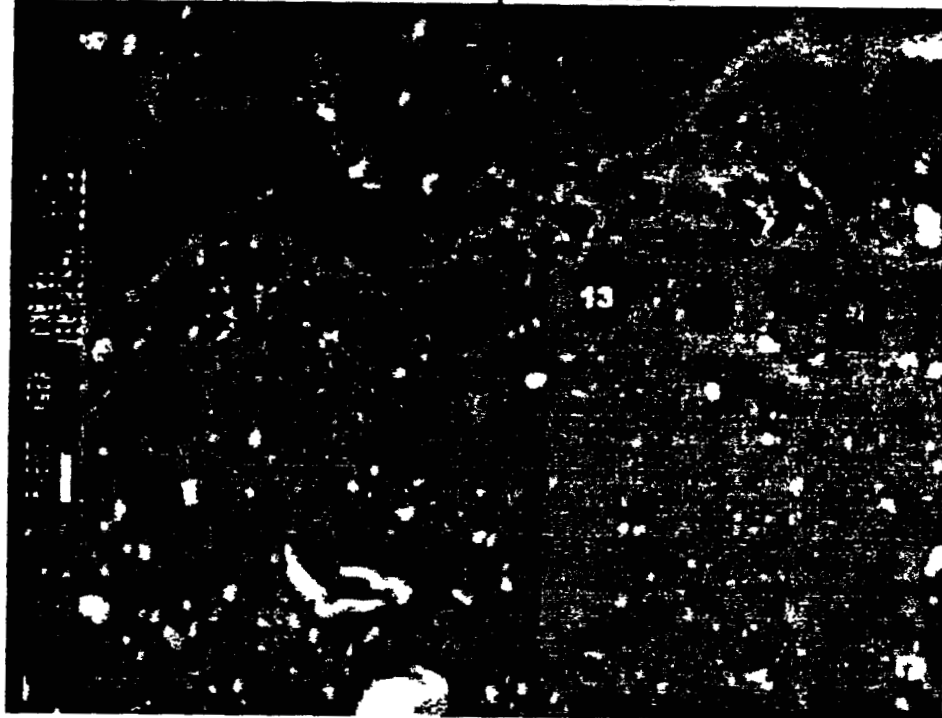
Figure 7c. TEM pictures of colloid samples. RF137N-3
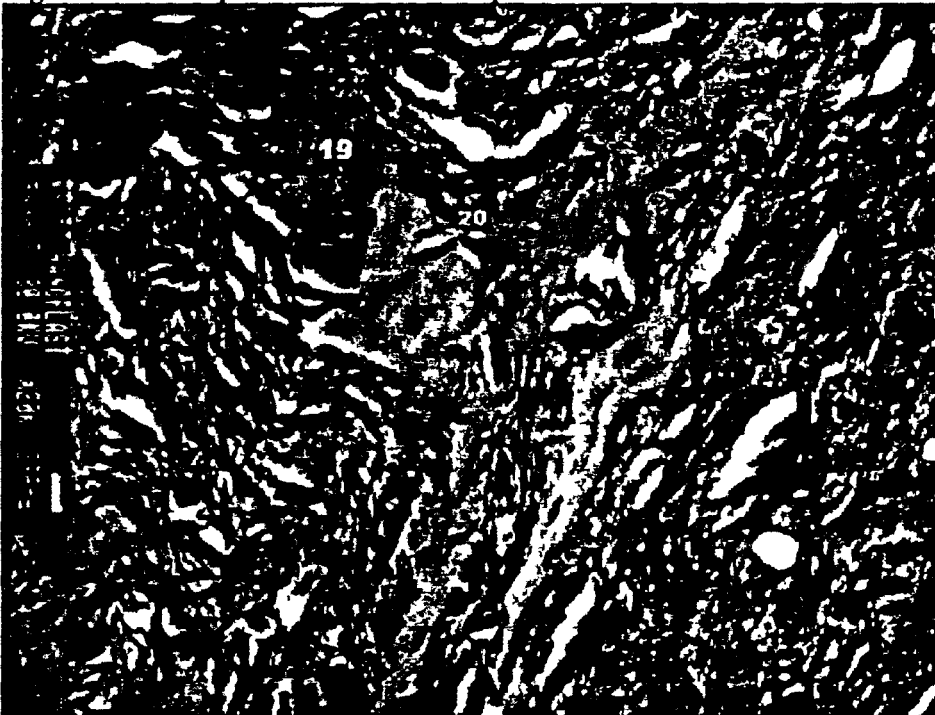

Figure 7d. TEM pictures of colloid samples RF137N-4

21
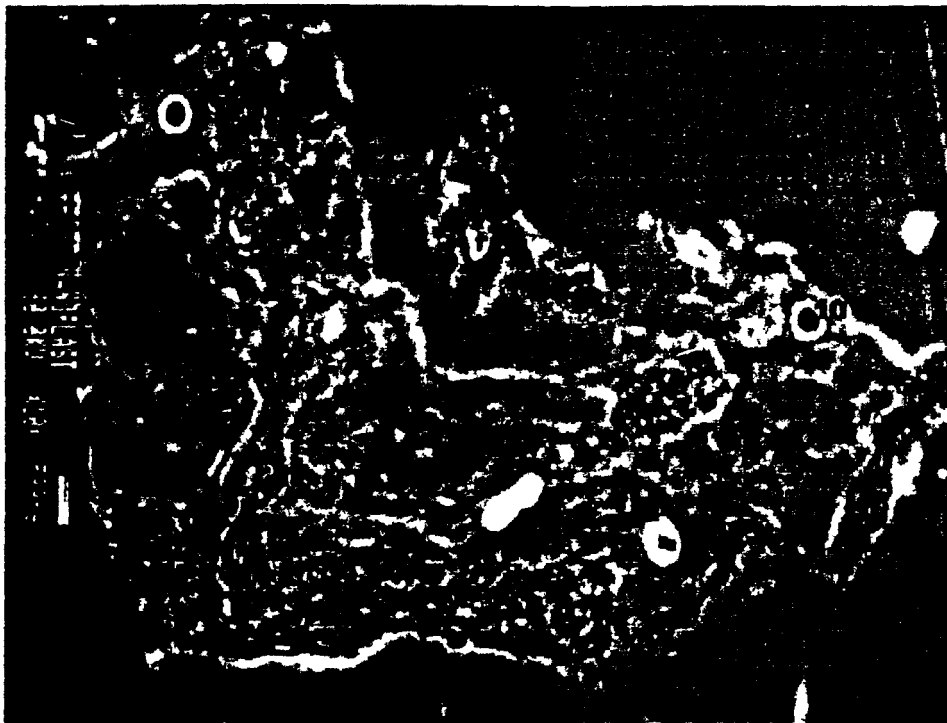
Figure 7e. TEM pictures of colloid samples. RF137N-5

Figure 7f. TEM pictures of colloid samples. RF137N-6

Figure 7g. TEM pictures of colloid samples. RF137N-7
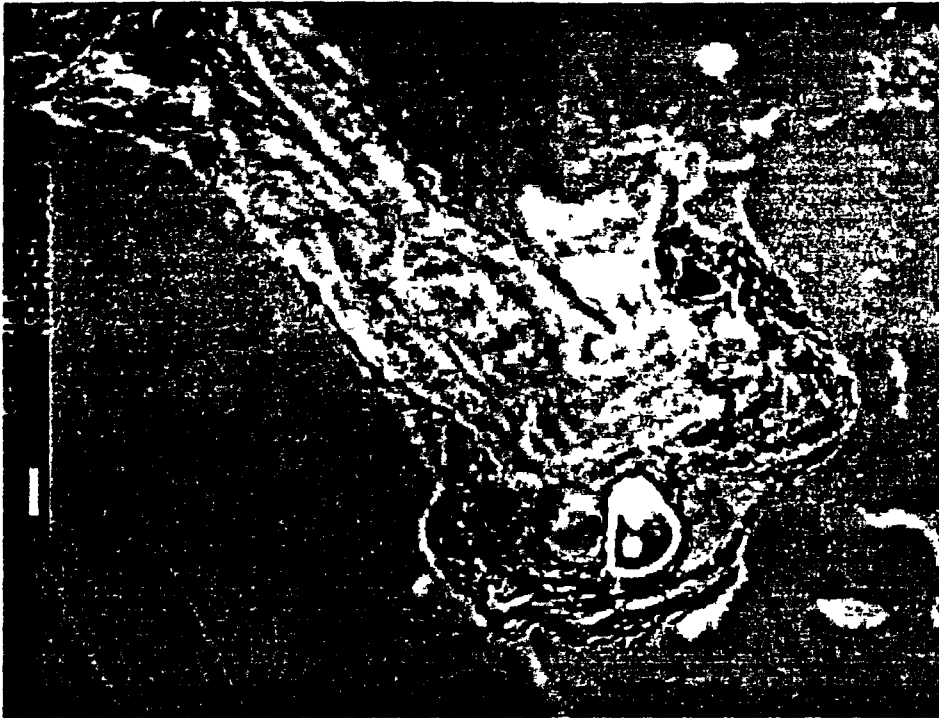
Figure 7h TEM pictures of colloid samples. RF157N-1
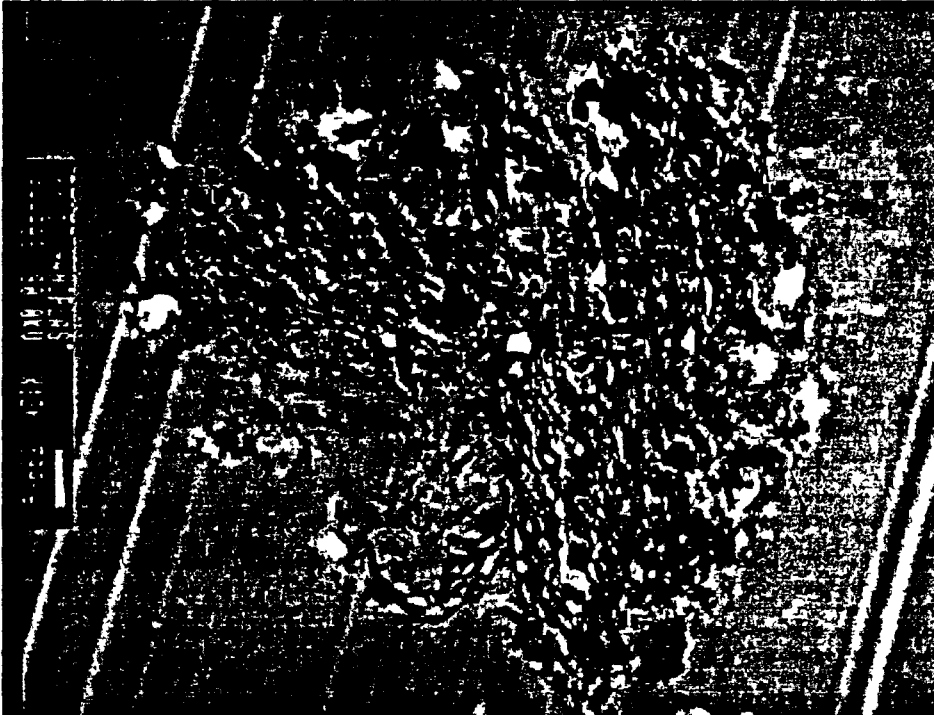
Figure 7ı TEM pictures of colloid samples. RF157N-2

24

Figure 7k TEM pictures of colloid samples. RF157N-3

Figure 7l. TEM pictures of colloid samples. RF157N-4
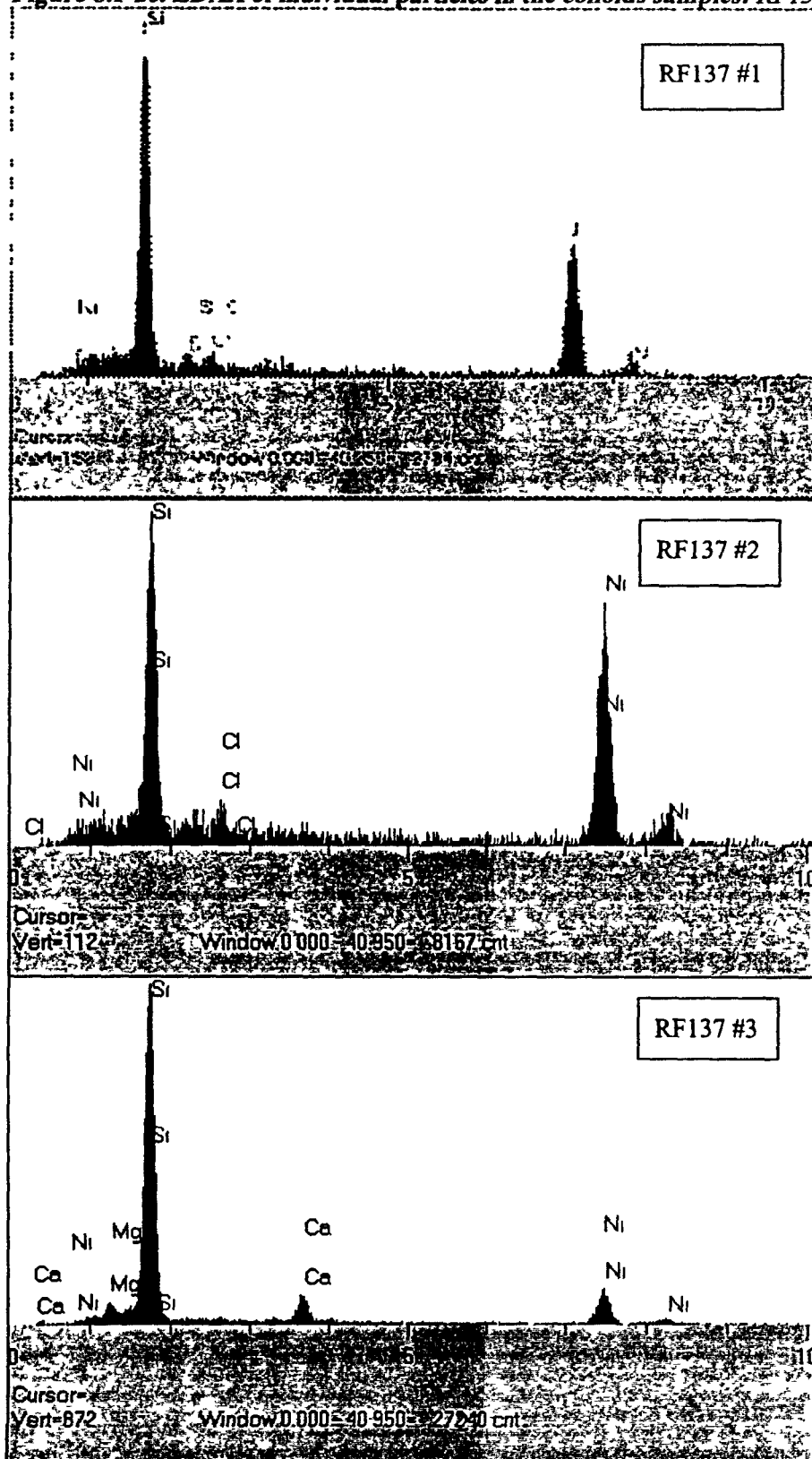
25

24

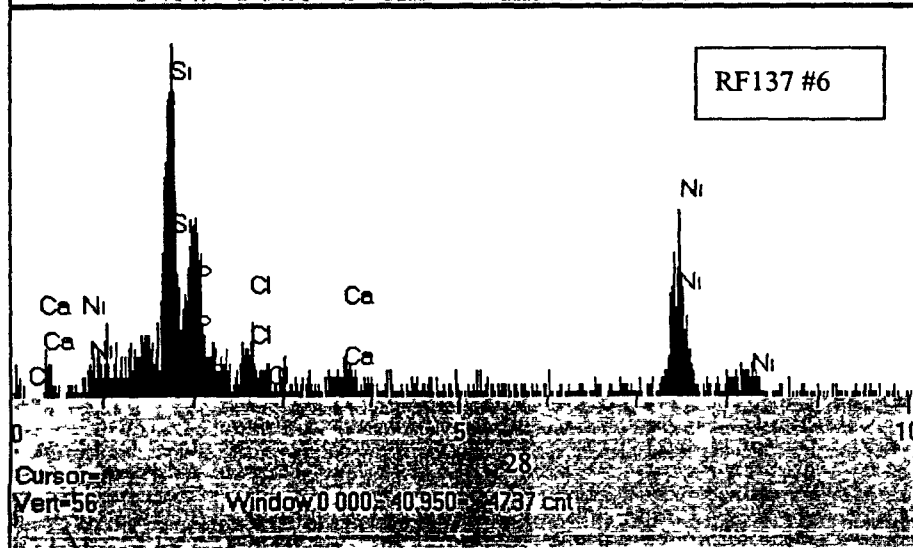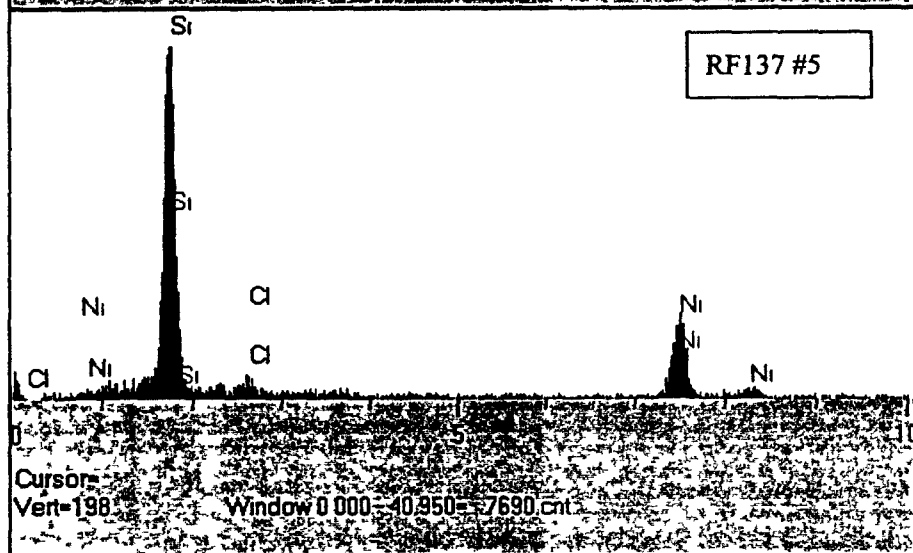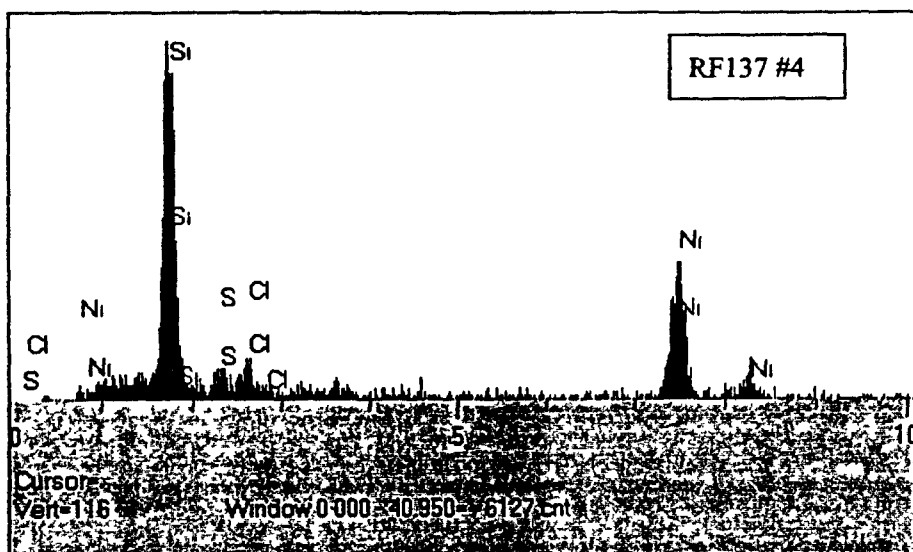**Figure 7m. TEM pictures of colloid samples RF157N-5**

Figure 7n TEM pictures of colloid samples RF157N-6

26

Figure 8.1-20. EDAX of individual particles in the colloids samples. RF137 Nanoplast

RF137 #1

RF137 #2

RF137 #3

RF137Nanoplast

RF137 #4

RF137 #5

RF137 #6

27

RF137Nanoplast

RF137 #7

RF137 #8

RF137 #9

28

RF137 #10

RF137 #11

RF137 #12

29

RF137Nanoplast

RF137 #13

RF137 #14

RF137 #15

30

RF137Nanoplast

RF137Nanoplast

RF137 #19

RF137 #20

33
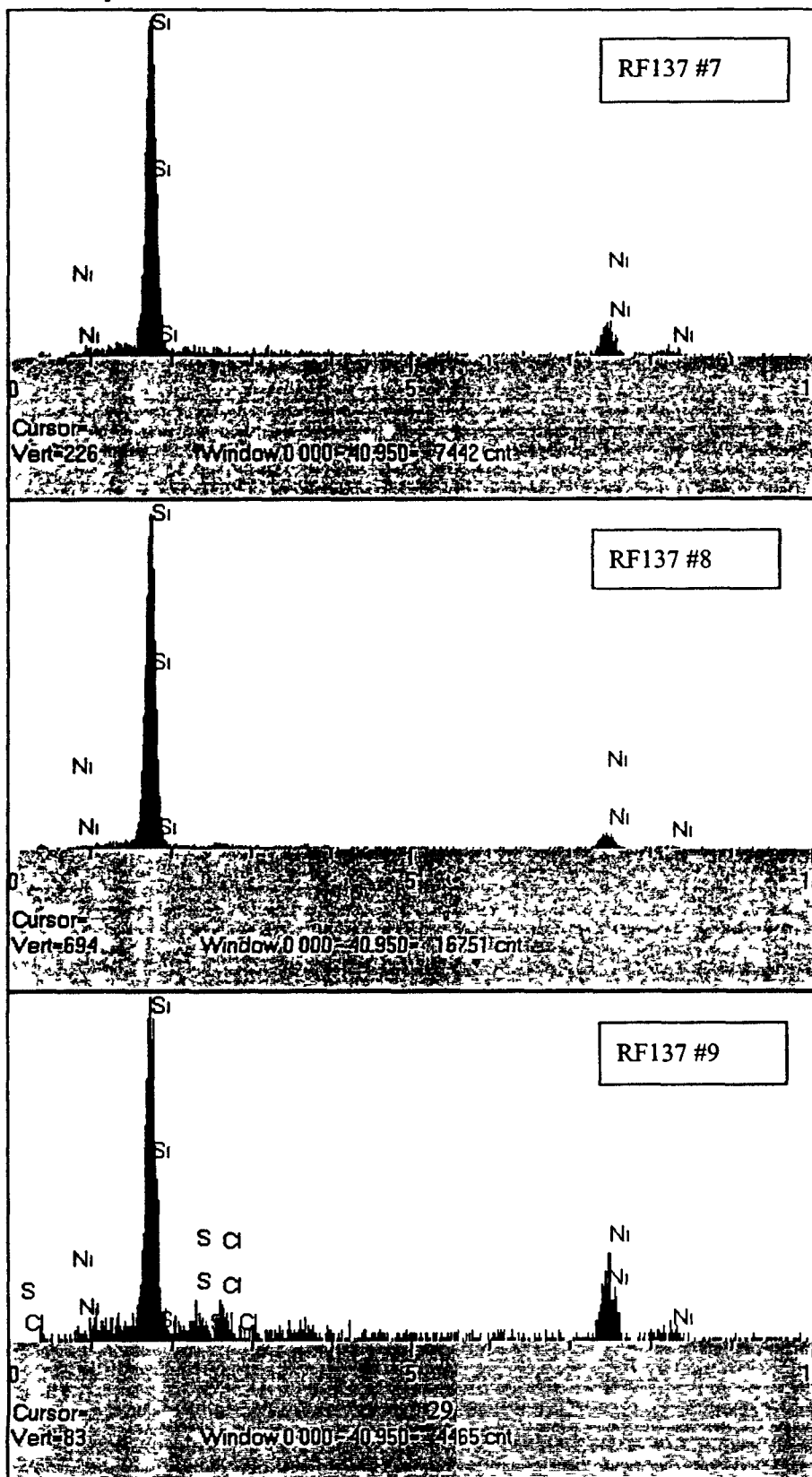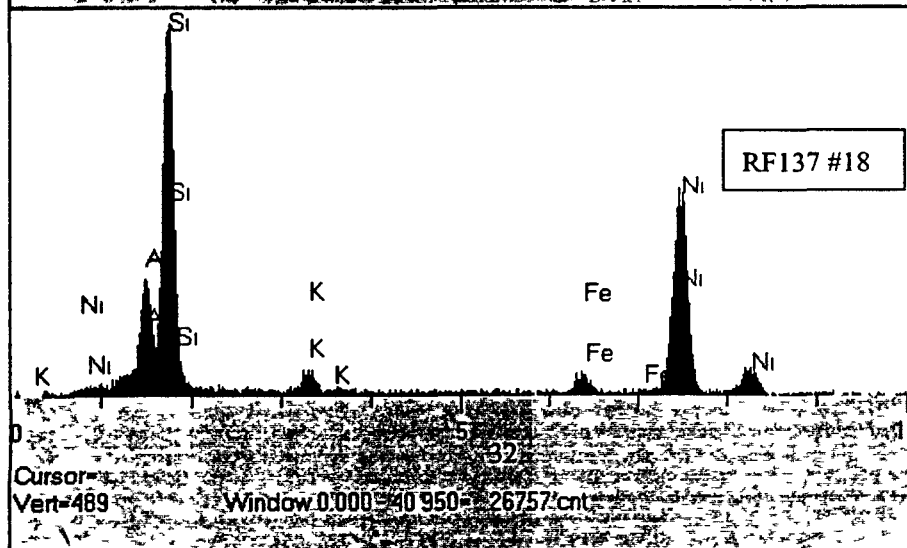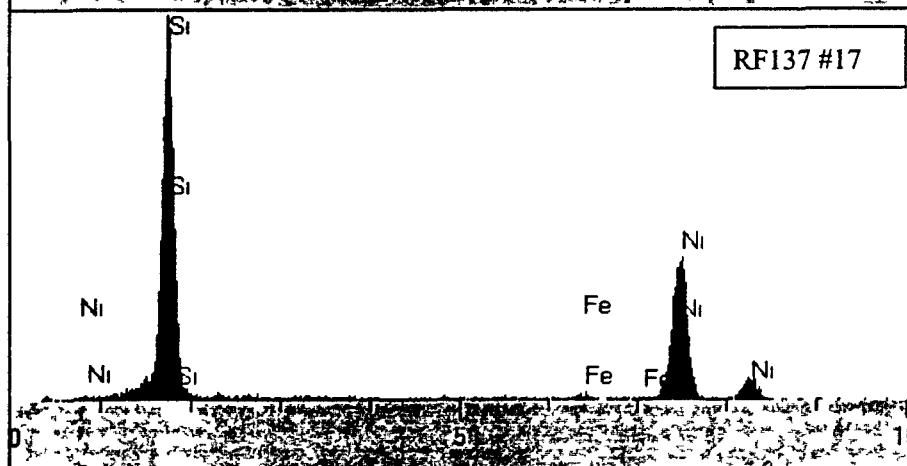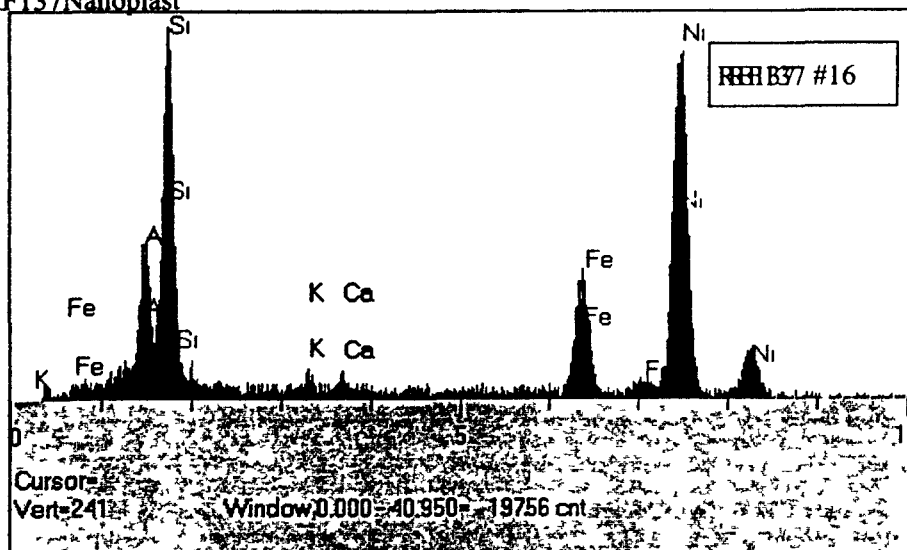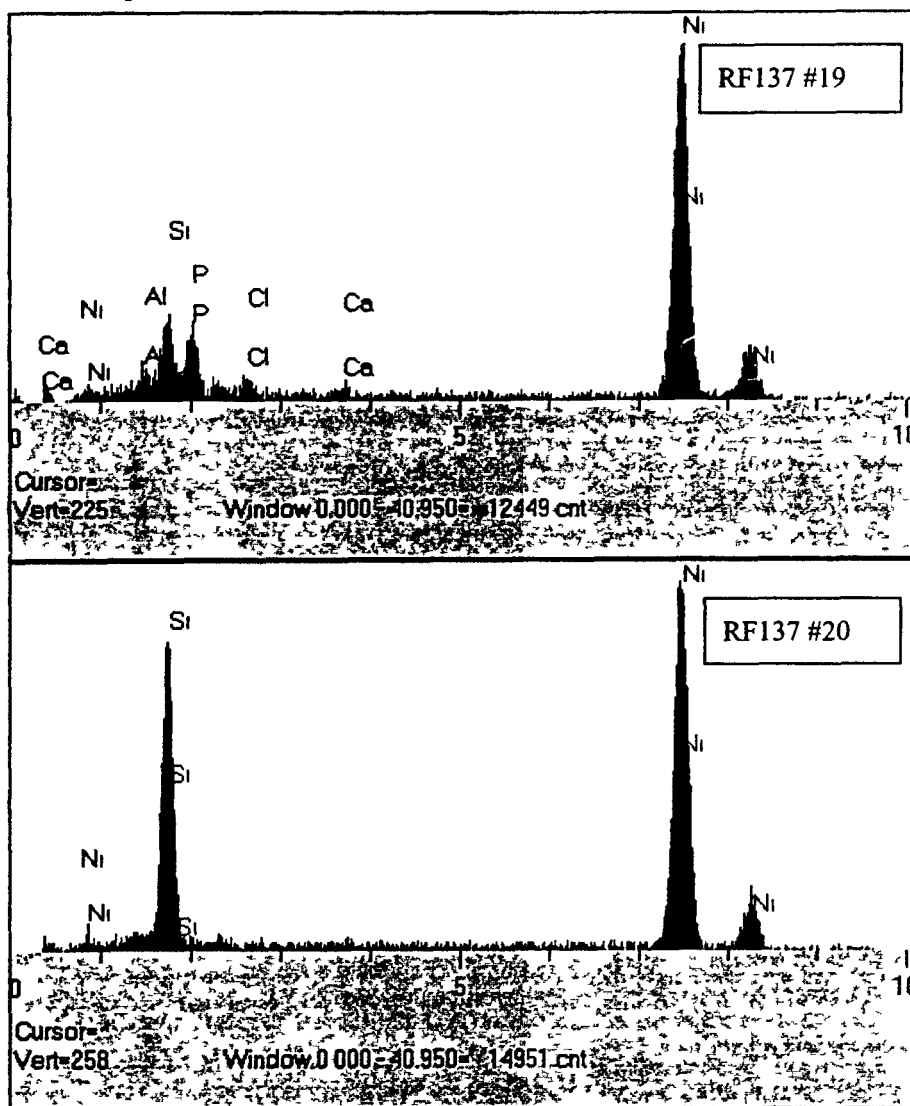
Figure 8.21-40. EDAX of individual particles in the colloids samples:
RF157Nanoplast

RF157 #1
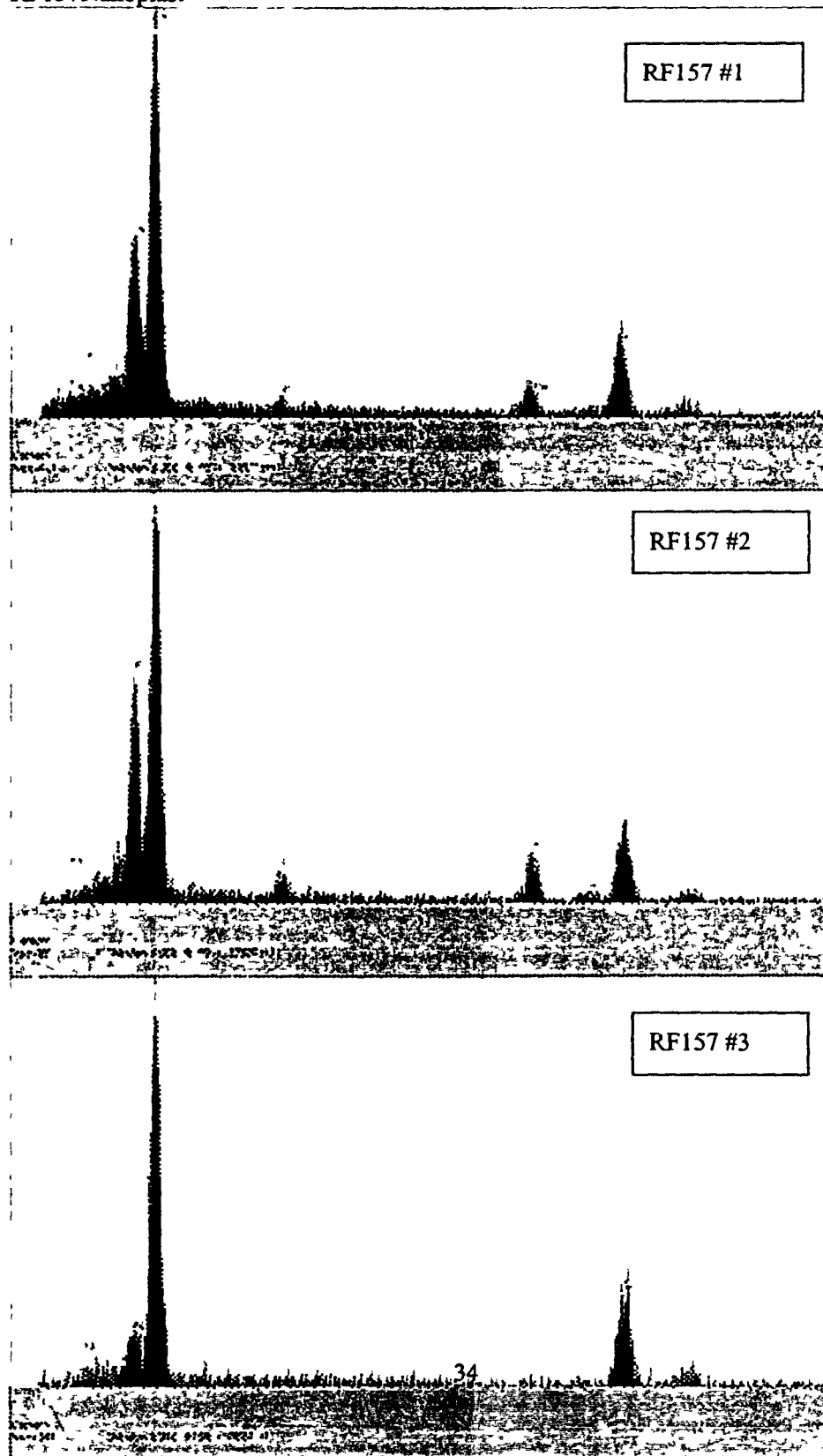
RF157 #2

RF157 #3

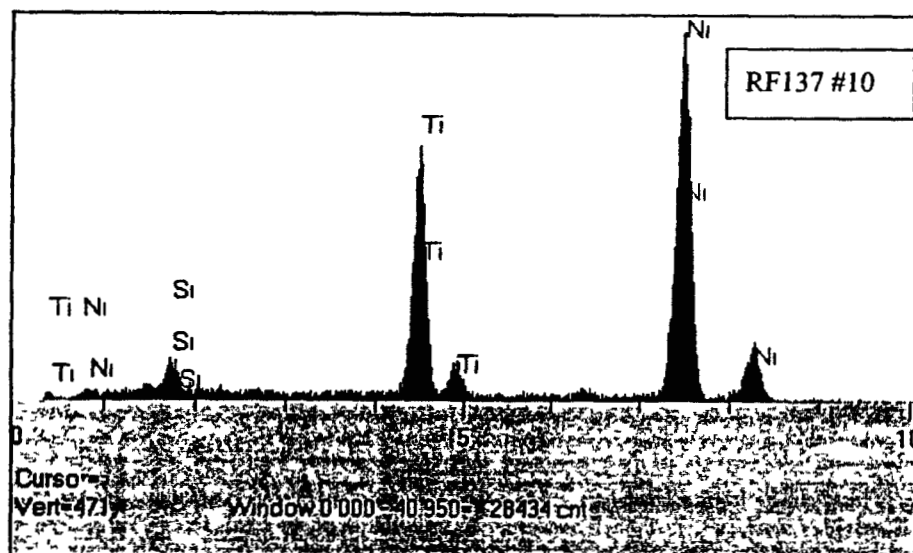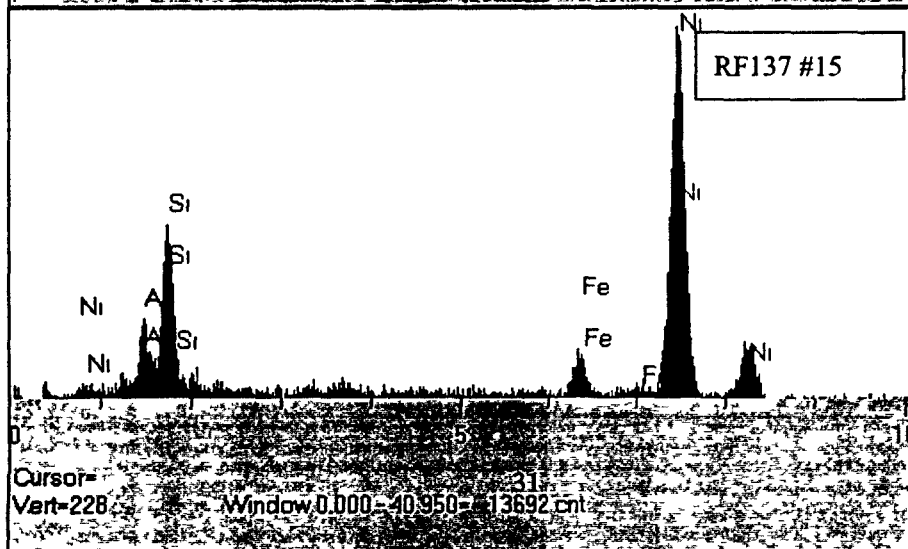RF157Nanoplast

RF157 #4

RF157 #5

RF157 #6

34
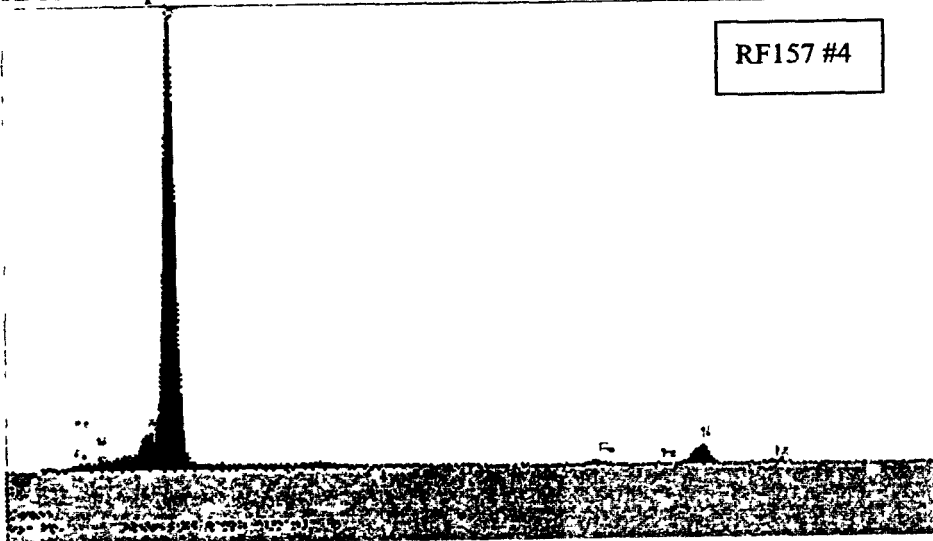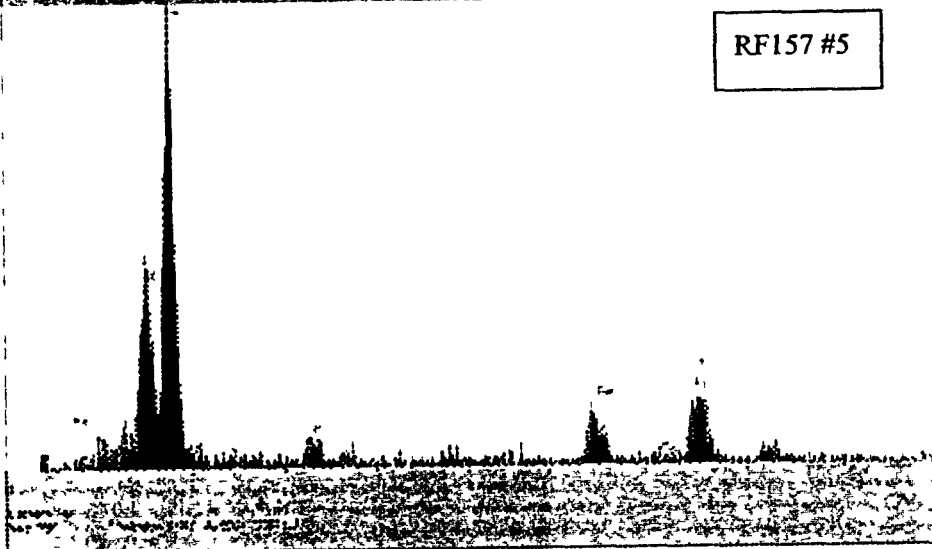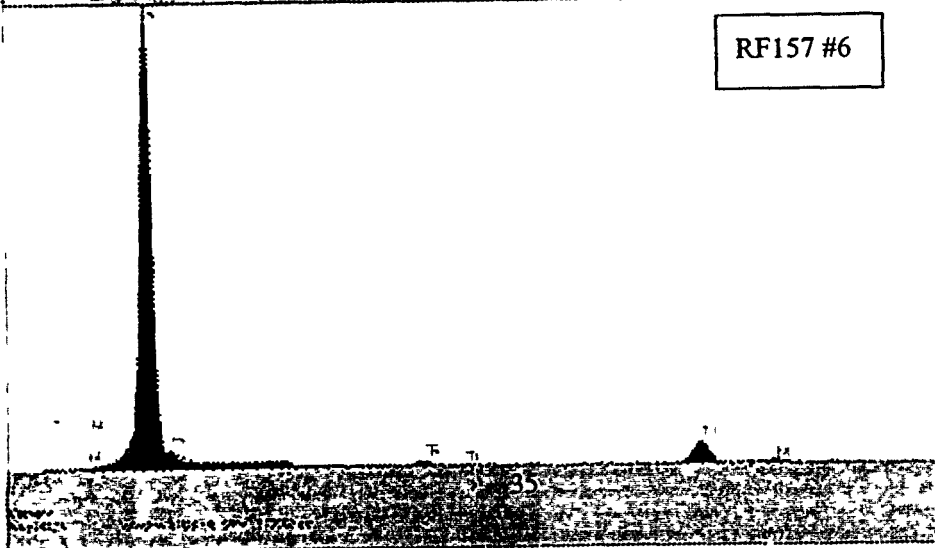
RF157 #9

RF157 #8

RF157 #7

RF157Nanoplast

35

RF157 #15

38

RF157 #14

RF157 #13

RF157Nanoplast

37
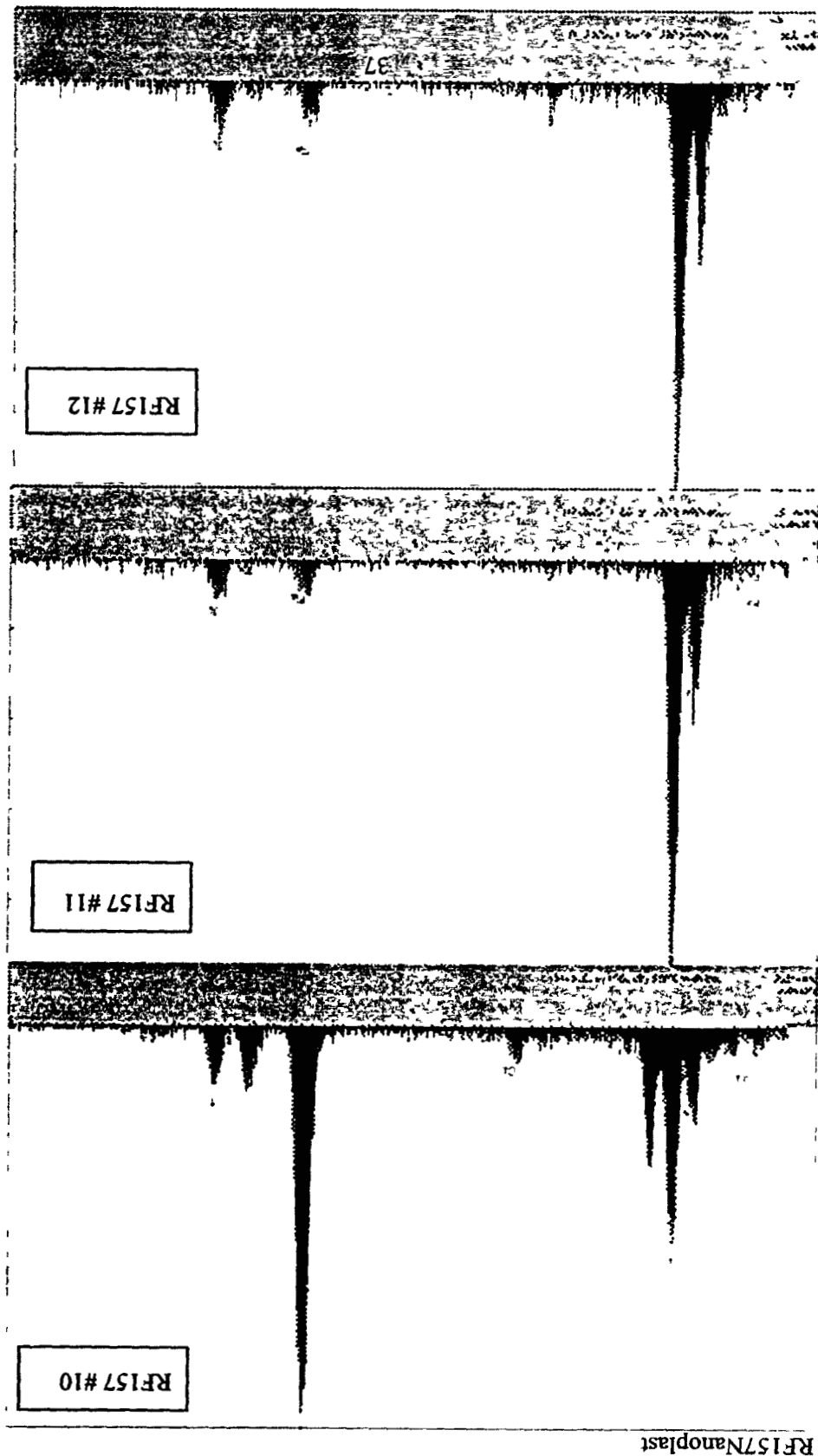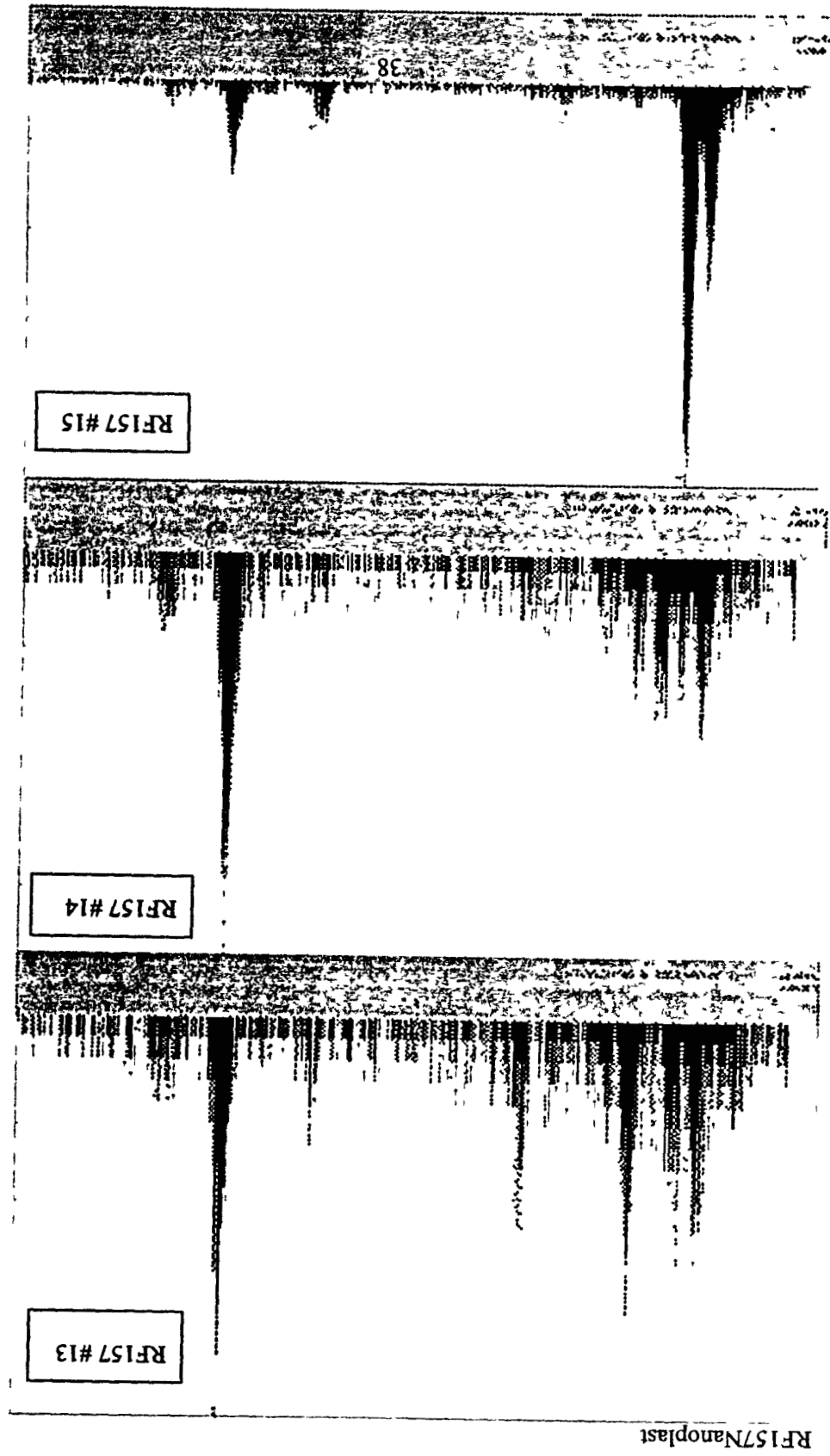
RF157Nanoplast

RF157 #16

RF157 #17

RF157 #18

89

38

RF157 #19

RF157 #20

RF157Nanoplast

39
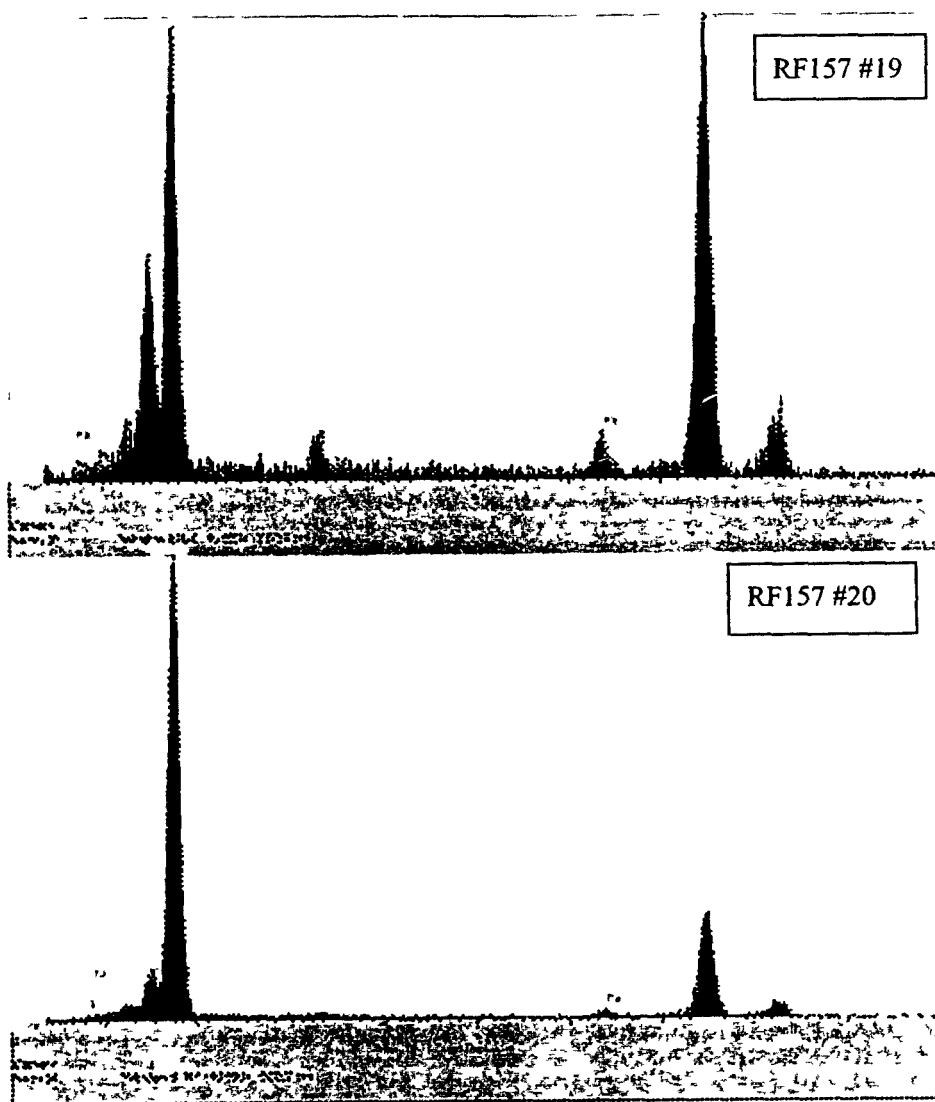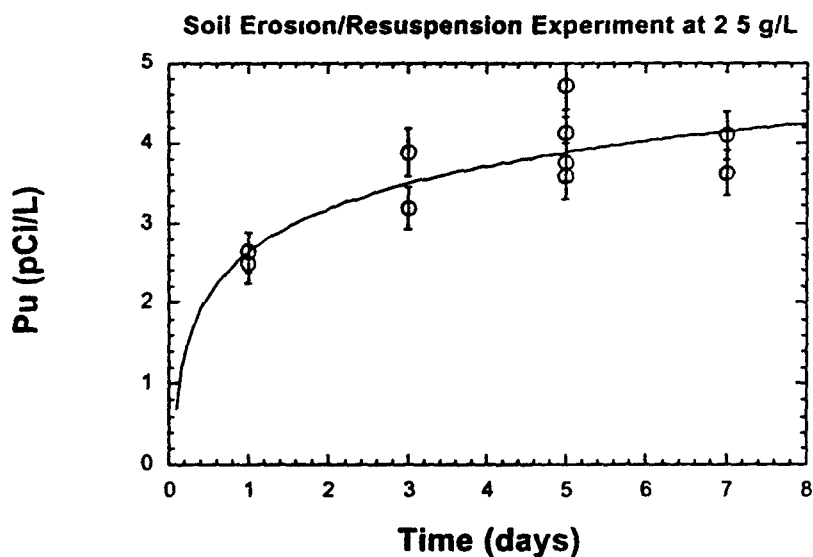
Figure 9. Kinetics of colloidal Pu remobilization during soil resuspension, with no humic acids added. DOC concentrations in solution, resulting from soil resuspension alone, ranged from 2.5 to 2.7 mg/L DOC.
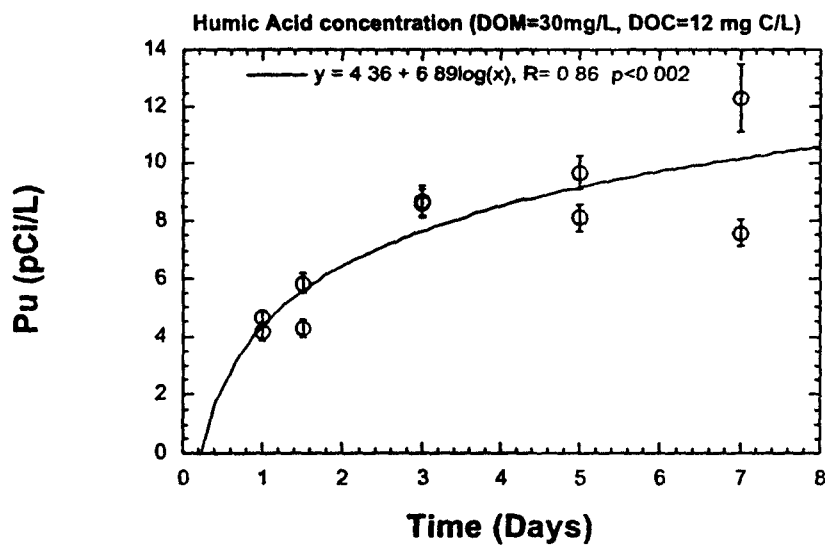
Figure 10. Kinetics of colloidal Pu remobilization during soil resuspension, with a constant amount of Aldrich humic acid added to the experimental solution

41
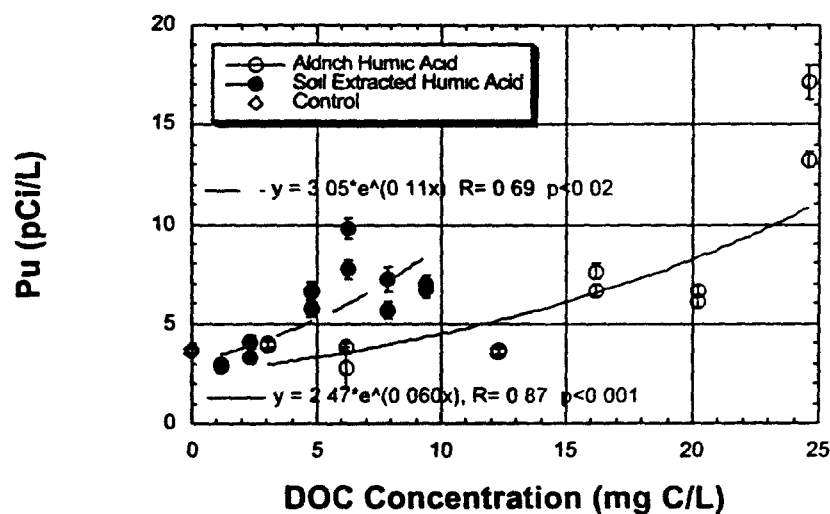
Figure 11. Comparison between Aldrich (41 % OC) and RFETS soil extracted (16 % OC) humic acid additions to the experimental solutions at resuspension times ranging from 3-7 days.
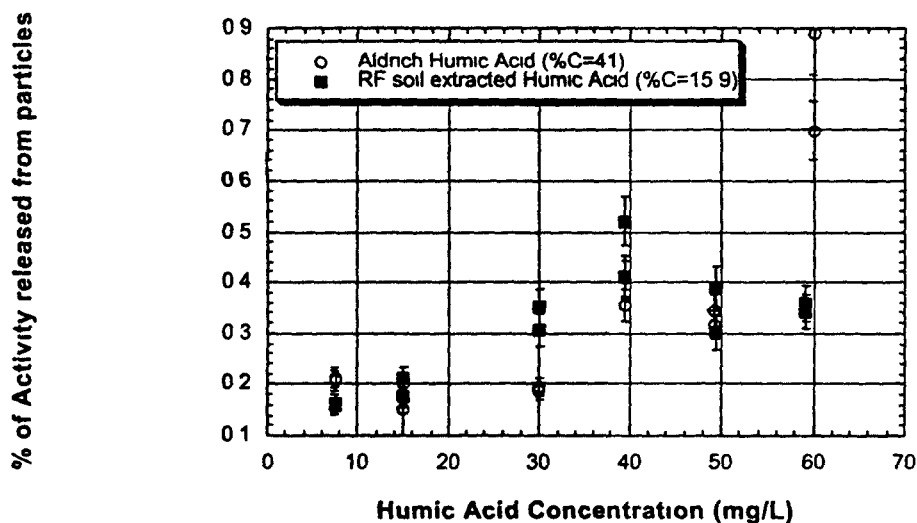
Figure 12. Percent Pu released from the application of different types of humic acids during 3-7 days of soil resuspension. Note, that humic acid concentration is given here as humic matter (DOM) concentration, while in Figure 9, it is in DOC concentration

41
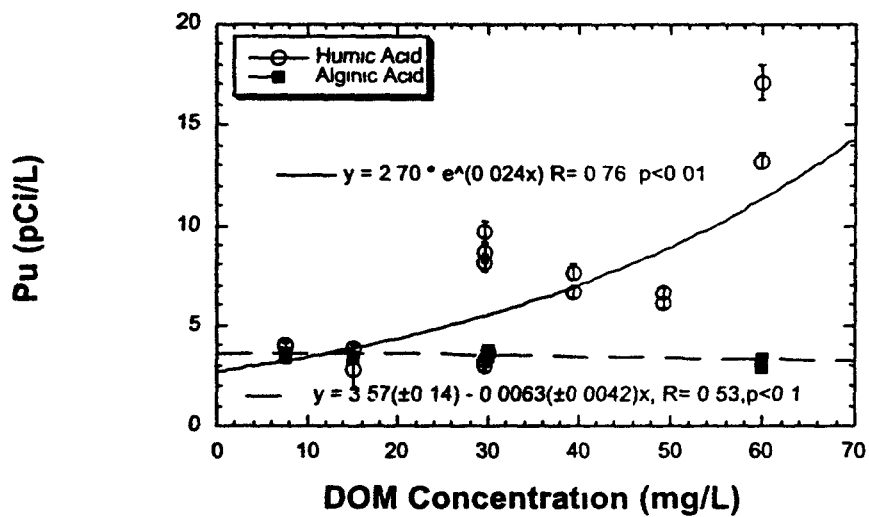
Figure 13. Comparison between Aldrich Humic Acid treated and Alginic Acid treated Pu remobilization experiments by soil resuspension/erosion.

42
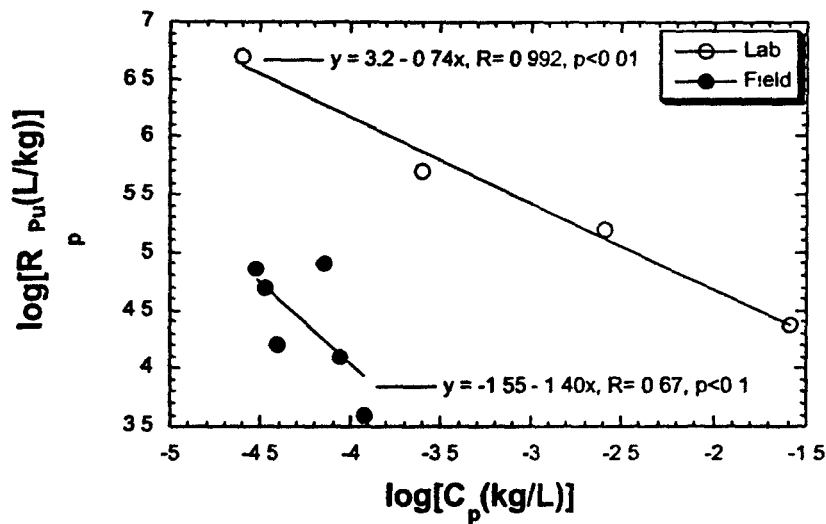
Figure 14. Relationship between particle concentration (Cp) and Phase partition coefficient (Rp) of Pu during soil resuspension experiments, after 4-5 days of particle resuspension, with no additional humic acid added. Laboratory results with RFETS soil taken near 903 Pad are also compared with field results from our water samples taken from GS03 and GS10 during 1998, 1999, and 2000.
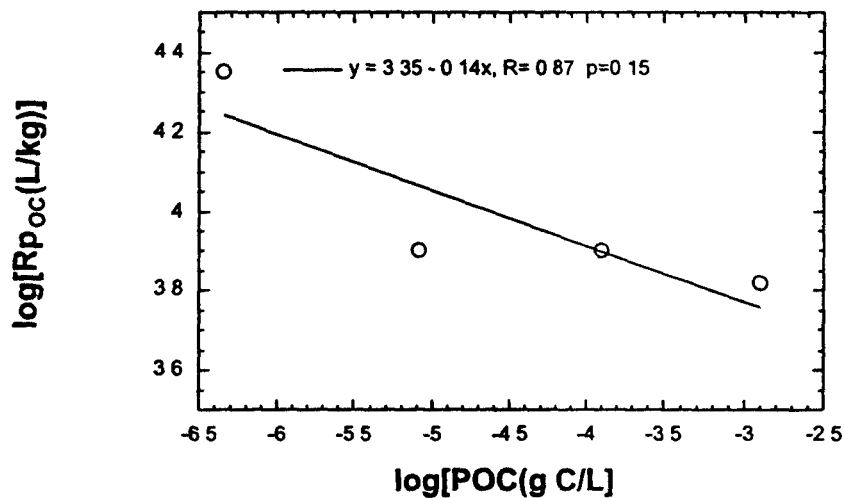
Figure 15. Relationship between particle concentration (Cp) and Phase partition coefficient (Rp) of organic carbon during soil resuspension experiments, after x days of particle resuspension, with no additional humic acid added.

43

Percentage of ^{239,240}Pu activities in the 0 45 micron filter passing
fraction with varying suspended soil concentrations

% of Activity released from particles

1 2

1

0 8

0 6

0 4

0 2

0

0 01   0 1   1   10   100

Suspended soil concentration (g/L)
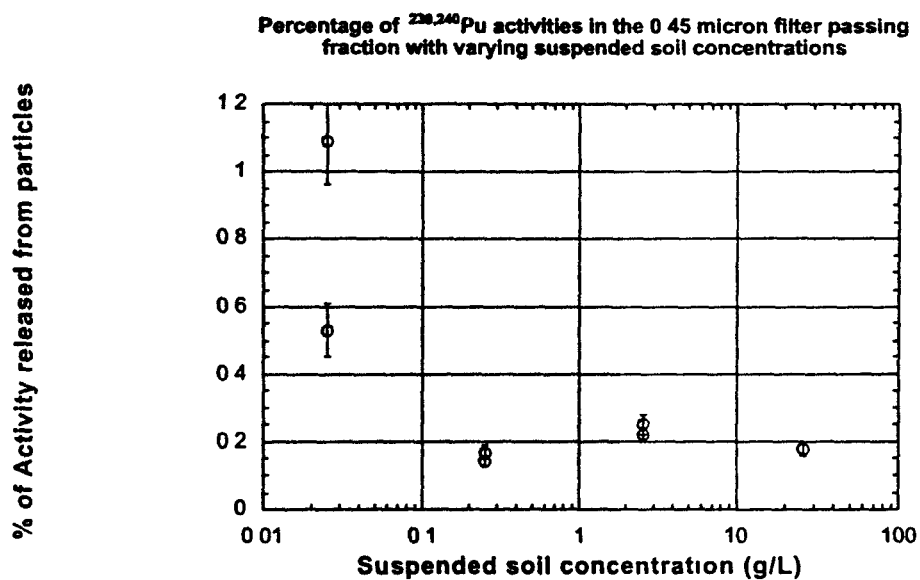
Figure 16. Fraction of soil-bound Pu which is released during simulated soil erosion and resuspension experiments, after 3-5 days of resuspension

44

Table 1. Summary of ^{239,240}Pu in duplicate samples A & B (SD = Standard deviation)

Sample ID	Fraction	A (pCi/l)	SD	B (pCi/l)	SD
4/27/00	Total	0 0588	0 0018	0 0503	0 0166
(Discharge)	TD	0 0225	0 0013	-	-
	>20μm	-	-	-	-
	0 5-20μm	-	-	-	-
	100K-R	0 0077	0 0007	0 0015	0 0005
	100K-U	0 0047	0 0006	0 0031	0 0007
	100K-Wash	0 0172	0 0006	0 0010	0 0005
	3K-R	0 0421	0 0119	0 0059	0 0008
	3K-U	0 0496	0 0095	0 0139	0 0012
	3K-Wash	-	-	0 0025	0 0006

Sample ID	Fraction	A (pCi/l)	SD	B (pCi/l)	SD
5/8/00	Total	0 0822	0 0034	0 1082	0 0043
(Storm)	TD	0 0142	0 0013	0 0070	0 0012
	>20μm	-	-	-	-
	0 5-20μm	0 0834	0 0021	0 0424	0 0013
	100K-R	0 0079	0 0006	0 0067	0 0007
	100K-U	0 0041	0 0007	0 0050	0 0012
	100K-Wash	0 0023	0 0005	0 0008	0 0005
	3K-R	0 0164	0 0012	0 0175	0 0014
	3K-U	0 0020	0 0006	0 0015	0 0007
	3K-Wash	-	-	-	-

U = ultrafiltrate (permeate), R = retentate, wash = cartridge wash solution - = tracer recovery <10%

Table 2. Summary of [241]Am data

Sample ID	Fraction	pCi/l	SD	pCi/l	SD
4/27/00	Total	0 0017	0 0008	0 003	0 001
(Discharge)	<0 5μm	-	-	-	-
	>20μm	-	-	-	-
	0 5-20μm	-	-	-	-
	0 5μm- 100kDa	0 0007	0 0004	-	-
	<100kDa	0 0022	0 0004	-	-
	100kDa-Wash	0 0065	0 0005	-	-
	0 5μm- 3kDa	0 0030	0 0008	-	-
	<3kDa	0 0030	0 0005	-	-
	3kDa-Wash	0 0067	0 0006	-	-

Sample ID	Fraction	pCi/l	SD	pCi/l	SD
5/8/00	Total	0 0672	0 0028	0 0768	0 0033
(Storm)	<0.5μm	0 0023	0 0003	0 0092	0 0009
	>20μm	-	-	-	-
	0 5-20μm	0 1377	0 0028	0 0406	0 0020
	0 5μm- 100kDa	0 0030	0 0005	0 0041	0 0004
	<100kDa	0 0023	0 0005	0 0030	0 0004
	100kDa-Wash	0 0031	0 0006	0 0020	0 0004
	0 5μm- 3kDa	0 0106	0 0009	0 0050	0 0004
	<3kDa	0 0018	0 0006	0 0006	0 0004
	3kDa-Wash	0 0009	0 0004	0 0008	0 0004

U = ultrafiltrate (permeate), R = retentate, wash = acid wash solution - = tracer recovery <10%

47

Table 3 . Suspended Particulate Matter (SPM, ≥0.45μm) Concentration.

Sample ID	Filter #	SPM (mg/l)	Average (mg/l)	SD	SD (%)
4/27/00-A	8	36 4	34 4	2 8	8 2
"	10	32 4			
4/27/00-B	5	28 8	27 5	1 9	7 2
"	6	25.2			
"	7	28 4			
5/8/00	1	53 6	72 4	16 5	22 8
"	2	84 4			
"	4	79 2			

48

Table 4. Particulate Organic Carbon (POC, ≥0.7μm) Concentration

Sample ID	Filter #	POC (mg-C/l)	Avg(mg/l)	SD	SD(%)
4/27/00	RF021	2 93			
"	RF022	3.29			
"	RF023	3 29	3 17	0 21	6 56
5/8/00	RF015	2 03			
"	RF016	1 98			
"	RF018	2 13	2 05	0 08	3 73

Table 5: Particulate Organic Nitrogen (PON, ≥0.7μm) Concentration.

Sample ID	Filter #	PON (mg-N/l)	Avg (mg-N/l)	SD	SD(%)
4/27/00	RF021	0 52			
"	RF022	0 57			
"	RF023	0 63	0 57	0 06	9 61
5/8/00	RF015	0.21			
"	RF016	0 21			
"	RF018	0 21	0 21	0 00	0 00

49

Table 6. C/N atomic Ratio of suspended particulate matter (≥0.45μm).

Sample ID	Filter #	POC (mg-C/l)	PON (mg-N/l)	C/N Ratio	Avg	SD
4/27/00	RF021	2 93	0 52	6 59		
"	RF022	3 29	0 57	6 75		
"	RF023	3 29	0 63	6 11	6 49	0 33
5/8/00	RF015	2 03	0 21	11 31		
"	2RF016	1 98	0 21	11 03		
"	3RF018	2 13	0 21	11 87	11 40	0 43

50

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Table 7. Dissolved organic carbon (DOC, <0.5µm) concentration.

Sample ID	Container #	DOC (ppm)	Avg (ppm)	SD (ppm)
4/27/00	A-1	4 232		
"	A-2	3 648	3 94	0 41
"	B-1	3 733		
"	B-2	3 453	3 59	0 20
5/8/00	A-1	5 094		
"	A-2	5 492		
"	A-3	11 92		
"	A-4	5 384		
"	A-5	5 413	6 66	2 94
"	B-1	5 431		
"	B-2	5 341		
"	B-3	5 082		
"	B-4	5 325		
"	B-5	5 237	5 28	0 13

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Table 8. Carbon and Nitrogen (%) in colloids, as average of duplicate measurements.

Sample ID	Description	Size	%C	%N	C/N
RF137	Discharge	3kDa–0 5μm	3 10	0 21	14 81
RF147	Discharge	3kDa–0 5μm	3 87	0 24	16 52
RF157	Storm	3kDa–0 5μm	11 94	0 600	19 88
RF167	Storm	3kDa–0 5μm	10 68	0 60	19 88

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Table 9. Trace Metal Concentrations in aqueous (4/27/00 and 5/8/00) and colloidal (RF137, RF147, RF157 and RF167) samples.

Sample ID	Subsample	Fe (ppb)	Al (ppb)	Mn (ppb)
Pond B5	total	45 55	35 24	70 72
	<0 5	26 13	23 34	38 5
	<0 5	26 92	24 55	42 37
	<0 5	26 57	25 84	41 21
4/27/00	A	40 31	15 81	35 14
	B	34 94	14 81	32 30
5/8/00	A	281 41	291 67	19 43
	B	279 67	290 43	21 87

Sample ID	Colloid size	Fe (mg/g)	Al (mg/g)	Mn (µg/g)
RF137	3kDa-0 5µm	0 15	0 07	86 05
RF147	3kDa-0 5µm	0 20	0 16	60 52
RF157	3kDa-0 5µm	10 30	13 32	131 41
RF167	3kDa-0 5µm	8 69	11 91	148 54

Table 10. Inorganic anion (Cl, NO3, HPO4, and SO4) concentrations.

Sample ID	Subsample	Cl (ppm)	NO3 (ppm)	HPO4 (ppm)	SO4 (ppm)
POND B5	4/26-1	319 03	86 08	0 88	46 74
	4/26-2	293 27	85 67	1 75	42 50
	4/26-3	308 55	85 55	1 0	36 06
	4/26-4	322 89	85 45	1 19	43 03
	4/27-5	314 26	83 47	1 06	37 63
	4/27-6	305 50			29 61
4/27/00	1	>250	7 83	-	61 78
	2	>250	7 71	-	
	3	>250	6 80	-	56 63
	4	>250	7 04	-	57 51
5/8/00	1	>250	28 20	-	7 87
	2	>250	28 35	-	7 93
	3	>250	28 25	-	7 77
	4	>250	28 04	-	7 74

\# NO_2 Concentrations are below detection limit (BD)

Table 11. Ancillary data taken at time of sampling by site personel.

Parameter	B5 4/26/00 10 45	B5 4/26/00 15 25	B5 4/27/00 9 10	GS03 4/27/00
pH	9 84	9 95	9 76	8 62
Temperature (°C)	15 3	16 5	13 8	17 6
SC (mS/cm)	1 444	1 391	1 264	1 172
DO(mg/L)	14 47	15 98	14 34	7 62
ORP				
Alkalinity(mg/L Ca)	110	112 5	100	131
EH				

Table 12. ^{239,240}Pu/^{241}Am ratios in dissolved, colloidal and particulate samples.

Date	Sample Name	Fraction	Pu/Am-1		Pu/Am-2		Pu/Am-Avg
4/27/00	Discharge	Total	34 59	16 31	16 77	7 86	25 68
		<0 5μm	-	-	-	-	-
		>20μm	-	-	-	-	-
		0 5-20μm	-	-	-	-	-
		0 5μm- 100kDa	11 00	6 36	-	-	-
		<100kDa	2 14	0 47	-	-	-
		100kDa-Wash	2 65	0 22	-	-	-
		0 5μm- 3kDa	14 03	5 45	-	-	-
		<3kDa	16 53	4 20	-	-	-
		3kDa-Wash	-	-	-	-	-
5/8/00	Storm event	Total	1 22	0 07	1 41	0 08	1 32
		<0 5μm	6 17	0 98	0 76	0 15	3 47
		>20μm	-	-	-	-	-
		0 5-20μm	0 61	0 02	1 04	0 06	0 82
		0 5μm- 100kDa	2 63	0 48	1 63	0 23	2 13
		<100kDa	1 78	0 49	1 67	0 46	1 72
		100kDa-Wash	0 74	0 22	0 40	0 26	0 57
		0 5μm- 3kDa	1 55	0 17	3 50	0 40	2 52
		<3kDa	1 11	0 50	2 50	2 03	1 81
		3kDa-Wash	-	-	-	-	-

WH=whole water, TD=total dissolved (i e , <0 5μm)

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Table 13. Phase distribution coefficients (R_p, l/kg or ml/g) of ^{239,240}Pu and ^{241}Am

Isotope	Sampling Date	Sample Name	C_p (mg/l)	R_D (ml/g)
^{239,240}Pu	4/27/00	Discharge	34	4 8 10^4
^{239,240}Pu	5/8/00	Storm event		
^{241}Am	4/27/00	Discharge	72	8 0 10^4
^{241}Am	5/8/00	Storm event		

C_p (mg/l) is the concentration of suspended particulate matter

5l

Table 14. Percent of total activity recovered during isoelectric focusing

pH	^{14}C RF137	^{14}C RF 157	^{59}Fe RF137	^{59}Fe RF 157
<3 9	47 79	52 90	2 50	1 96
3 9	17 15	20 84	0 45	1 26
4 4	7 67	5 08	0 70	1 29
4 8	4 05	4 09	0 86	1 29
5 3	4 79	3 01	6 49	1 90
5 8	4 79	2 19	2 22	1 97
6 3	3 52	1 34	2 95	3 42
6 8	2 04	2 48	7 29	9 29
7 3	2 44	1 55	9 95	15 99
7 8	1 31	2 17	3 40	11 86
8 3	1 31	0 62	59 06	7 35
8 7	1 64	1 88	3 55	8 74
>8 7	1 51	1 88	0 57	33 69

Table 15: Phase speciation results using a 1kDa stirred cell ultrafiltration after resuspending 0.5g RFETS soil in 190ml of filtered tap water.

	Fraction	pCi/L	SD	%	pCi/L	SD	%
Control	<0 45	2 812	0 253		5 547	0 409	
	0 45-1kDa	2 649	0 324	94 21	3 572	0 378	64 40
	<1kDa	1 011	0 209	35 97	0 896	0 153	16 15
	Filter leach	0 004	0 003	0 14	0 013	0 006	0 23
	Sum	3 665	0 386	130 33	4 481	0 408	80 77
Humic Acid (29 6mg/L)	<0 45	7 141	0 507		6 428	0 444	
	0 45-1kDa	4 565	0 430	63 94	4 819	0 506	74 98
	<1kDa	0 152	0 071	2 13	0 368	0 110	5 72
	Filter leach	0 043	0 010	0 60	0 074	0 019	1 15
	Sum	4 760	0 436	66 66	5 261	0 518	81 85